United States Court of Appeals
for the
District of Columbia Circuit

TRANSCRIPT OF RECORD

BRIEF FOR APPELLANT

United States Court of Appeals

FOR THE DISTRICT OF COLUMBIA CIRCUIT

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For the
District of Columbia Circuit

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No. 14,704

PURDUE RESEARCH FOUNDATION,

Appellant,

v.

ROBERT C. WATSON,
Commissioner of Patents,

Appellee.

414

APPEAL FROM THE JUDGMENT OF THE UNITED STATES DISTRICT COURT FOR THE DISTRICT OF COLUMBIA

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STATEMENT OF QUESTIONS PRESENTED

(1) Whether a claim in a patent application meets the statutory requirement of Title 35, United States Code, Section 112 to particularly to point out and distinctly claim the novel compound of applicants' invention, when such claim defines the otherwise patentable compound in terms of its color, crystalline structure, approximate melting point, chemical behavior, solubility and approximate molecular formula.

(2) Whether a claim particularly points out and distinctly claims the inventive compound when such claim defines the compound, not only in terms of its color, crystalline structure, approximate melting point, chemical behavior, solubility and approximate molecular formula, but also recites a process whereby it can be prepared.

(3) Whether a claim describing a compound in terms of its "fingerprint" characteristics, each of which separate characteristics applies or could apply to other compounds, adequately defines the inventive compound when the evidence shows that the sum-total of the characteristics can apply to only one compound.

(4) Whether a claim containing the expression "melting point of about 355" and "approximate molecular formula $C_{10}Cl_{10}O$" meets the requirements of 35 USC 112, when the specification and the level of knowledge of those skilled in the art to which the invention pertains, serve to place adequate limits on the modifying words "about" and "approximate" and when compounds having such high melting points and such molecular formulas are very rare.

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UNITED STATES COURT OF APPEALS
For The District Of Columbia Circuit

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PURDUE RESEARCH FOUNDATION,
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APPEAL FROM THE JUDGMENT OF THE UNITED STATES
DISTRICT COURT FOR THE DISTRICT OF COLUMBIA

BRIEF FOR APPELLANT

JURISDICTIONAL STATEMENT

This is an appeal under Section 1291 of Title 28, United States Code, 65 Stat. 726, from the judgment of the United States District Court for the District of Columbia, dismissing a complaint against the Commissioner of Patents, defendant-appellee, in an action brought under Section 145 of Title 35, United States Code, 66 Stat. 803, in which a decree was sought authorizing the Commissioner to grant letters patent of the United States for the invention specified in the claims of United States patent application Serial 238 186, filed 1951 July 23 by

Earl T. McBee and Jack S. Newcomer, entitled "Composition of Matter." Appellant is the assignee of the entire right, title and interest in and to said application.

STATUTES INVOLVED

The only statute involved is Title 35, United States Code, section 112, which reads:

> "The specification shall contain a written description of the invention, and of the manner and process of making and using it, in such full, clear, concise, and exact terms as to enable any person skilled in the art to which it pertains, or with which it is most nearly connected, to make and use the same, and shall set forth the best mode contemplated by the inventor of carrying out his invention.
>
> "The specification shall conclude with one or more claims particularly pointing out and distinctly claiming the subject matter which the applicant regards as his invention.
>
> "An element in a claim for a combination may be expressed as a means or step for performing a specified function without the recital of structure, material, or acts in support thereof, and such claim shall be construed to cover the corresponding structure, materials, or acts described in the specification and equivalents thereof."

STATEMENT OF CASE

This case is concerned solely with the question of whether certain claims in a patent application directed to admittedly novel, useful chemical compounds, properly define the otherwise patentable and inventive subject-matter.

Counsel for the parties agreed at the trial that claims 5, 7 and 8 should stand or fall with claim 6; and that if either claim 6 or 13 is allowed, the other should not be allowed (JA 12, 13).

At the time the application was filed, and even now, applicants did not know the exact structure of their novel chemical compound (which if known would automatically provide a name for the compound). They did ascertain and know many of the most important chemical and physical properties of the compound and used these as the basis for their claims. The chemical compound involved is unusual in that it exists in several forms because it is very hygroscopic (i.e. it picks up water when exposed to the atmosphere) and it forms hydrates depending upon how much water it has picked up. It is very difficult to tell when the compound exists in the anhydrous form (without any water) or in one of the several possible hydrated forms (depending upon just how much water, a compound of hydrogen and oxygen, the compound has picked up). This difficulty was recognized by applicants, and in fact the application as originally filed contained the following first paragraph:

> "The present invention relates to a novel composition of matter, more particularly to a chemical compound believed to have the approximate molecular formula $C_{10}HCl_{10}O_2$. While the exact elemental composition and structure of the new compound are not precisely known at the present time, sufficient of its physical and chemical properties have been determined to allow its positive identification and production by any one of several procedures." (JA 43)

The molecular formula is a determination of how many atoms of what elements go into the whole compound, and is determined by chemical analysis of a sample.

Subsequently applicants found that they had based their original analysis on a sample of the compound which contained some water, rather than one which was entirely dry, so they revised the approximate molecular formula to $C_{10}Cl_{10}O$, which differs from the original molecular formula by HO, which is about one water molecule, H_2O.

Claim 6 reads as follows:

> 6. A composition of matter which has the following characteristics: it is a white crystalline solid having a sealed tube corrected melting point of about 355 degrees Centigrade, it behaves as a weak organic acid, is very soluble in polar organic solvents, substantially insoluble in non-polar organic solvents, and has the approximate molecular formula $C_{10}Cl_{10}O$.

Claim 13, which contains all the descriptive language of claim 6, plus an additional phrase telling one way the compound is prepared, reads as follows:

> 13. A composition of matter which has the following characteristics: it is a white crystalline solid having a sealed tube corrected melting point of about 355 degrees Centigrade, it behaves as a weak organic acid, is very soluble in polar organic solvents, substantially insoluble in non-polar organic solvents and has the approximate formula $C_{10}Cl_{10}O$ and is preparable from a compound having a melting point of approximately 146-147 degrees Centigrade, formed by the reaction of hexachlorocyclopentadiene with chlorosulfonic acid, by treatment with alcoholic alkali and acidification and dehydration.

Claims, of the general type of claims 6 and 13, are not objectionable *per se* as a type of claim, and are often allowed by the Patent Office.

The claims were objected to mainly because of the use of certain qualifying language, i.e. "melting point of *about* 355 degrees," and "*approximate* molecular formula $C_{10}Cl_{10}O$" (which appear in Claims 6 and 13) and in the use of "is prepar*able* by" (rather than "is prepar*ed* by" in claim 13).

Claim 6 defines the compound in terms of the following characteristics: color [white]; form [crystalline solid]; melting point [about 355 degrees Centigrade]; chemical behavior [behaves as a weak organic acid]; solubility in various solvents; and molecular formula [approximate molecular formula $C_{10}Cl_{10}O$]. From this information, any chemist by a simple mathematical calculation, will also know the molecular weight, and the percentage of carbon (C), chlorine (Cl) and oxygen (O) present in the compound. (JA 23) Chemical Handbooks which contain data about thousands of chemical compounds, list as the more important identifying physical constants for solids: name, formula, molecular weight, crystalline form, color, melting point and solubilities. (JA 107, 110) Except for the name, all of these important identifying characteristics are listed in the claim.

The Patent Application

The application Plaintiffs Exhibit 1, contains the following pertinent disclosure about each of the characteristics mentioned in the claim:

A. As to "behaves as a weak organic acid":

(1) "The compound --- is a ---- solid, ----, having properties of a weak organic acid, ---" (JA 44)

(2) "The compound ---- behaves as does a weak organic acid in that it gives an indefinite end-point with phenolphthalein, but a definite end-point with thymolphthalein." (JA 46)

(3) "The compound ----- reacts with basic reagents to form salts." (JA 49)

B. As to "having a ---- melting point of about 355 degrees centigrade":

(1) "The compound-----, when in the anhydrous state, [has] ---- a ----- melting point ----- of about 355 degrees centigrade." (JA 44)

(2) "This novel composition of the present invention exists in the anhydrous form as well as in the form of various hydrates. The compositions, both hydrous and anhydrous, are odorless white solids which melt ---- at about 355 degrees centigrade." (JA 46)

(3) "The hydrate, the anhydrous form, and mixtures of the hydrate with the anhydrous form, all give the same melting point --- of approximately 355 degrees centigrade." (JA 47)

(4) When the compound was prepared by the method of Example 2, drying of samples of the same compound by various methods gave the compound with 1.75 percent water, 2.13 percent water, 5.82 percent water, 0.15 percent water, and 0.19 percent water. All of the materials were found to melt within the range 350-357 degrees centigrade and none depressed the melting point of the others. (JA 51)

(5) The compound, when prepared as in Example 3, had a melting point of 351 - 353 degrees centigrade. (JA 52)

(6) The compound, when prepared as in Example 5, had a melting point of 350-355 degrees centigrade. It did not depress the melting point of the materials prepared in Examples 2 and 3. (JA 53)

(7) The compound, when prepared as in Example 6, melted at 339-359 degrees centigrade, and was considered to be a hydrate. (JA 54)

(8) The compound, when prepared as in Example 7, melted at 350-352 degrees centigrade. (JA 54)

(9) The compound, when prepared as in Example 8, melted at 350-357 degrees centigrade, and did not

depress the melting point of previously prepared samples when mixed melting points were taken. (JA 55)

C. As to "is very soluble in polar organic solvents":

(1) The compound is "very soluble in polar organic solvents such as acetone, methyl alcohol, ethyl alcohol, acetic acid ---" (JA 46)

D. As to "is --- substantially insoluble in non-polar organic solvents":

(1) The compound "is insoluble in non-polar organic solvents such as carbon tetrachloride and benzene." (JA 46)

E. As to "has the approximate molecular formula $C_{10}Cl_{10}O$":

(1) The application as filed said that the claimed compound was "believed to have the approximate molecular formula $C_{10}HCl_{10}O_2$" and also that "the exact elemental composition...are not precisely known at the present time ----" (JA 43)

(2) There was never any doubt about the chlorine [Cl]. Thus: "The compound, when in the anhydrous state,---containing about 70 percent chlorine---" (JA 44); and, "The presence of ten chlorine atoms in the compound of approximate formula---is indicated by the analysis presented in Table I supra, as well as by the fact that two chlorine atoms are removed from the compound $C_{10}H_2Cl_{12}O_3S$ when it is converted to the compound----. These data are shown in Table III." (JA 49)

(3) The molecular formula was converted by amendment to $C_{10}Cl_{10}O$. (JA 64-66)

(4) The Examiner accepted the correction of the molecular formula, and adopted it in a claim which he suggested would probably be allowable. (JA 73)

F. As to "and is preparable from a compound having a melting point of approximately 146-147 degrees centigrade, formed by the reaction of hexachlorocyclopentadiene with chlorosulfonic acid, by treatment with alcoholic alkali and acidification and dehydration" (which appears in claim 13):

This application discloses that the compound "may be produced by any one of a considerable number of methods". JA 44-45 generally states at least four different processes including that mentioned in claim 13, all of which are specifically illustrated in Examples 2, 3, 5, 6, 7 and 8. (JA 51-55)

The Prosecution Within The Patent Office

The Examiner, in Paper No. 7, suggested a claim as to which he said at JA 73:

> "The following claim would probably be allowable: A composition of matter which has the following characteristics: it is a white crystalline solid having a sealed tube corrected melting point of about 355 degrees centigrade, it behaves as a weak organic acid, is very soluble in polar organic solvents, substantially insoluble in non-polar organic solvents, and has the approximate formula $C_{10}Cl_{10}O$ and is prepared by the heating of $C_{10}H_2Cl_{12}O_3S$ with a member of the group consisting of potassium hydroxide, methanol, potassium iodide, and activated carbon and subsequent dehydration."

Applicants did not adopt that claim, which was a "product by process" claim. Instead they continued to assert claims closely based on the quoted claim, and after final rejection appealed unsuccessfully to the Patent Office Board of Appeals, the sole question presented being that of adequacy of definition of that [claimed] compound in accordance with 35 U. S. Code (1952 Ed.) Section 112. (JA 99)

The Trial

At the trial before the District Court, the sole witness was James S. Sconce, a skilled chemist employed by The Hooker Electrochemical Company (JA 20). His testimony supplied the requisite interpretive background of the chemist skilled in the art of organic chlorine compounds and showed the unusualness of certain of the physical characteristics of the claimed compound.

Thus he testified as follows:

A. As to "having a ----, melting point of about 355 degrees centigrade":

(1) That Table I of the application (JA 48) which shows a melting point range, doesn't cover just one compound, but a compound and its hydrates (JA 20).

(2) How a chemist determines melting point and what points are significant (JA 22).

(3) That for compounds melting at lower temperatures, e.g. 20-50 degrees centigrade, the melting point can be determined very accurately to a hundredth of a degree, but that when the melting point is very high, e.g. 355 degrees centigrade, then the accuracy of a melting point determination is not as great and "temperature is a much wider figure, two degrees, three degrees or four degrees, is quite significant" (JA 22).

(4) That in the book "Organic Chlorine Compounds" by Huntress published in 1948, which lists all organic chlorine compounds known as of about 1946 (JA 31) there was no compound containing C, Cl, O or C, Cl, O, and H,

which contained from 68-73 percent chlorine, that had a melting point within 50 degrees of 355 degrees centigrade; that the nearest compound containing any of those elements was pentachloropropionic acid, $C_3HO_2Cl_5$ which melted at 200-215 degrees centigrade. (JA 22-23)

(5) That standard Handbooks such as Lange's "Handbook of Chemistry" and the Chemical Rubber Publishing Co.'s "Handbook of Chemistry and Physics", each of which lists well over 7,000 organic compounds, do not list any compound melting at 355 degrees centigrade; that The Handbook of Chemistry and Physics lists only 11 compounds melting between 350 and 360 degrees centigrade, and none of these contain chlorine; that Lange's Handbook lists only 9 compounds melting between 341 and 360 degrees centigrade, none of which contains chlorine. (JA 23-24)

(6) That standard Handbooks have charts which give the melting point ranges of organic compounds (JA 24, 107, 109) which chemists use as a standard procedure.

(7) That most organic compounds melt at lower temperatures, e.g. in the 50-70 degrees centigrade area, where for a 5-8 degree centigrade range as many as 200 compounds will be listed; that "It is clear that in compounds that melt at 300 degrees centigrade and higher, there are very, very few listed and known to the chemists." (JA 24)

(8) That "The melting point of a solid-----[is] the most used physical properties for the characterization of organic compounds" (JA 24, 111) and that our

handbooks list melting points as one of the most important and practical factors in identifying compounds.

B. As to "behaves as a weak organic acid":

(1) He explained the various ways in which the compound (which is not an acid) behaves like a weak organic acid, which are listed in the specification. (JA 24, 25, 35)

C. As to "soluble in polar organic compounds" and "substantially insoluble in non-polar compounds":

(1) He explained that chemists have a definite understanding as to which solvents are considered polar or non-polar based on their dipole moment. (JA 26, 35)

D. As to "has the approximate molecular formula $C_{10}Cl_{10}O$":

(1) That molecular formula is of significance even where the word "approximate" is used in its definition; he told the analytical procedures used to determine carbon, hydrogen, oxygen and chlorine; that the accuracy of analytical methods is complicated in compounds of this sort and by the purity of the original compound. (JA 26)

(2) That the compound can pick up moisture from the air to form a hydrate and "that was one of the reasons that the original formulas, as supplied, contained hydrogen because water is nothing more than H_2O, which are two hydrogens and one oxygen. (JA 27)

(3) That the molecular weight is calculable from the molecular formula, viz. the atomic weight of carbon (C) is 12; of chlorine (Cl) is roughly 35.5; of

oxygen is 16, so you multiply each atomic weight by the subscript, and add them (JA 11) to get the total molecular weight which is about 493; that "The significance of this 493 molecular weight is quite important. Now, we would expect to determine molecular weight accurately in that range to maybe five or six, or if the true molecular weight is 493, we would be satisfied with a molecular weight determination of 490 or 496. I don't think it would be much more reliable than that." In a lower range the determination would be much more accurate. (JA 27)

(4) That the atomic weight of hydrogen (H) is one, so that in analyzing for hydrogen you are hunting for 1 in 493; and that in seeking an additional oxygen, it was 16, applied against 493. (JA 27)

E. As to Identification of the Claimed Compound:

(1) That from his experience and knowledge, the combination of characteristics set forth in claim 6, accurately defines a single chemical compound, and only one. (JA 27)

(2) That it could be easily determined by use of a mixed melting point determination whether or not any other compound infringes (JA 38-39) the claimed compound.

(3) That a manufacturer could tell, without preparing the claimed compound, whether or not a new compound infringed. (JA 40)

The Exhibits

Aside from the Exhibits discussed above, "The Encyclopedia of Chemistry" shows the behavior of a weak organic acid (JA 114); and how chemists use the mixed melting point method to identify an unknown compound, by mixing a sample of the unknown compound with the known compound, and seeing if the melting point is different from that of the known compound itself (JA 115); that solvents are divided into two classifications based on polarity, i.e. polar solvents and non-polar solvents (JA 116).

STATEMENT OF POINTS

1. The court erred in finding that the melting point of "about 355 degrees centigrade" was not sufficiently accurate to make it an identifying characteristic.

2. The court erred, in not holding the claims specifying the melting point of "about 355 degrees centigrade", as particularly pointed out and distinctly claiming the invention against the background of the evidence that: such a melting point is so high as to be most uncommon among organic compounds; there are no compounds known containing carbon, chlorine and oxygen, or carbon, chlorine, oxygen and hydrogen that contain from 68 to 73 percent chlorine (as does the compound here claimed) which have a melting point within 50 degrees of 355 degrees centigrade.

3. The court erred in not finding that the melting point of "about 355 degrees centigrade" defines a significant identifying "finger print" characteristic having a clear, and restricted meaning to a chemist.

4. The court erred in finding that the phrase "approximate molecular formula $C_{10}Cl_{10}O$" could only have significance as indicating that the formula may be varied by a whole number of any one or more of the three kinds of atoms indicated, and that the phrase is meaningless as an item of identification of the claimed chemical compound.

5. The court erred in not finding that the slight change in the molecular formula as originally disclosed involved only a question as to how much hydrogen (H) and Oxygen (O), as water, (H_2O) of hydration, was or was not present in the base $C_{10}Cl_{10}O$ molecule.

6. The court erred in finding that the molecular formula $C_{10}Cl_{10}O$ is not derivable from the application as originally filed.

7. The court erred in not considering that the molecular formula gives to a chemist the molecular weight and percentage of chlorine, and that these are further identifying characteristics inherently contained in the claim language.

8. The court erred in finding that the claims do not specify the criterion of behavior to be ascribed to "behaves as a weak organic acid".

9. The court erred in not considering the definitions within the specification itself as providing a clear and definite meaning to the terms "behaves as a weak organic acid", "is very soluble in polar organic solvents," and "substantially insoluble in non-polar organic solvents."

10. The court erred in giving any consideration to the fact that the application as filed did not identify the chemical compound as a ketone.

11. The court erred in not considering the specification as the lexicon for the claims, in making its findings.

12. The court erred in finding that the characteristics of the claimed compound as set forth in claim 6 and in claim 13, which were referred to by plaintiff as the compounds "finger prints", do not, when taken cumulatively, adequately and distinctly describe the claimed compound.

13. The court erred in not considering the evidence that the particular characteristics stated in the claims include all those commonly given in chemical handbooks as identifying characteristics of chemical compounds, save for the name of the compound which is even now unknown.

14. The court erred in not finding that the claims are sufficiently clear and definite so a rival chemist could tell with reasonable certitude whether a substance he might make would infringe.

15. The court erred in not considering the evidence that in this area of organic chemistry it is impossible to be as precise as in some other areas, and that identifying characteristics such as those of the claims are accepted by chemists working in this area as sufficiently definite for their purpose.

16. The court erred in not considering the additional characteristic of claim 13 detailing one of the several ways of making the compound set forth in the specification, as rendering the claim clear and definite.

17. The court erred in concluding that claims 5-8 and 13 do not meet the requirements of 35 U.S.C. 112.

18. The court erred in concluding that plaintiff is not entitled to a patent containing any of the claims 5-8 and 13 of McBee et al. patent application serial 238 186.

19. The court erred in concluding that protection in product-by-process claims must be limited to compositions produced by the process referred to in the claims, under the particular facts of the instant case.

SUMMARY OF ARGUMENT

1. The claim language must be construed in the light of the specification and the level of knowledge of chemists skilled in the art of organic chlorine compounds to whom it is addressed, bearing in mind that the certainty required in patents is not greater than that which is reasonable, having regard to the subject matter involved, and that the degree of definiteness necessary to satisfy the law varies in accordance with the facts and circumstances of each case.

2. The claimed compound is here described by its identifying properties, some of which, i.e. melting point and molecular formula, are expressed in terms allowing for some variation from the exact (355 degrees and $C_{10}Cl_{10}O$) numerical quantities set forth in the claims.

The extent of this variation is easily ascertainable from the specification. This latitude is necessary because the hygroscopic nature of the compound and its exceptionally high melting point lead to a higher analytical margin of error than is present in many other compounds.

3. The numerical expressions of the particular properties of melting point and molecular formula are very unusual, so that each is of great identifying significance, even allowing for the permissible variation.

4. The chemical art normally identifies unknown compounds by comparing their properties with known compounds, and the properties enumerated in the claims include all the major identifying properties used.

5. Each of the properties enumerated in the claims has a clear and definite meaning to a chemist.

6. Considered separately, any one of the identifying characteristics recited by the claims might conceivably not exclude all other possible compounds having such selected individual properties - but, taken collectively, the recited characteristics do absolutely identify one single substance. It is the cumulative effect of the several recited properties, even allowing for some variation in the melting point and molecular formula, which narrows the field to one compound meeting such claims.

7. The claims are sufficient so that any chemist could tell, without undue effort, whether or not a "new" compound infringes.

ARGUMENT

The inventive subject-matter of the involved McBee et al. application is novel, useful and otherwise patentable if the claims are sufficient to "particularly point out and distinctly claim" the invention, as required by Title 35, United States Code, Section 112.

Claims are not to be considered in a vacuum and the following few quotations from cases involving analogous questions will be helpful in properly construing the statutory provision involved and establishing the proper climate to be applied, thus avoiding the errors of the lower court:

In Lever Bros. Co. v. Procter and Gamble Mfg. Co. et al (CCA 4), 139 F. 2d 633, 639, the Court, faced with determining whether claims to a soap and a soap-making process were sufficiently clear and definite, upheld them saying:

> "In all important legal instruments, the use of words with precision and exactness is surely a consummation devoutly to be wished. In few, if any, of these instruments is this quite so true as in the specifications and claims of a patent. Weasel words have no place there. But the federal statute hardly requires even an approximation of perfection. Semantics is not an exact science. And surely it is more difficult to obtain the desired precision and exactness in the description of an industrial process than in the description of a machine. The words of Bodman's specifications and claims must be judged in the light of the vocabulary available to a soapmaker.
>
> "We are fully conscious of 'the public interest sought to be safeguarded by the patent statutes, and so frequently present but so seldom adequately represented in patent litigation.' Mr. Justice Jackson, in the recent case of Muncie Gear Works v. Outboard Marine and Manufacturing Co., 315 U. S. 759, 768, 62 S. Ct. 865, 870, 86 L. Ed. 1171. We cannot forget, though, that the purpose of the patent statutes is also to stimulate invention and that inventors should not be deprived of the fruits of their labors by too technical a nicety in the requirement that inventions must be adequately described to indicate their real scope and their true content. As Circuit Judge Soper said, speaking for this Court, in Procter and Gamble Manufacturing Co. v. Refining, Inc., 4 Cir., 135 F. 2d 900, 906:
>
> "'***It was not necessary or practicable for Clayton to specify the exact degree of emulsion breaking temperature or the precise length of time needed for the substantial neutralization of every quality of oil that might be subjected to refinement; and the evidence shows that the information given was entirely adequate to persons skilled in the art. ***

"'There are many situations in the practice of the arts in which specific directions are properly omitted from claims of patents because greater definition is either impracticable or is unnecessary to inform the art, and would serve only unduly to limit the scope of the invention or to invite evasion by those who desire wrongfully to misappropriate the substance of the invention.'

"See, also, Minerals Separation, Ltd. v. Hyde, 242 U. S. 261, 270, 271, 37 S. Ct. 82, 61 L. Ed. 286; Eibel Process Co. v. Minnesota and Ontario Paper Co., 261 U. S. 45, 65, 66, 43 S. Ct. 322, 67 L. Ed. 523; Smith v. Snow, 294 U. S. 1, 7, 55 S. Ct. 279, 79 L. Ed. 721. See, too, the opinion of Circuit Judge Swan (quoted above) in Franc-Strohmenger and Cowan v. Arthur Siegman, Inc., 2 Cir., 27 F. 2d 785, 786.

"In the light of these observations, and the holdings in these cases, we venture the observation that the District Judge imposed too high a standard on Bodman's use of such terms as "closed", "continuous" "pressure", "plastic or semi-fluid", "texture", "characteristic" and "similar". Without attempting to discuss just what we regard as the precise connotation of each of these terms, we feel that, read in their context in Bodman's claims and specifications, these terms could be readily understood and successfully followed by experienced makers of soap.

"Germane, here, is Bodman's uncontradicted testimony (quoted above) that ordinary workmen in soap found no difficulty in understanding and following these terms used in his claims. We cannot expect in a soap-process patent (and it would be unreasonable to require) terms with the precise exactitude and the clear cut accuracy found in the nomenclature of pure mathematics. We are dealing with a practical, present field of industry, not with a visionary far-off Utopia. It would be, we think, altogether unreasonable to require strictly that Bodman describe his process only in words with a keen, razor-like connotation."

The Court of Custom and Patent Appeals, in Application of Hudson, 205 F. 2d 174, 177, in a case wherein the question was whether a boiling point limitation was properly expressed in a claim, applied the following standard:

> "The certainty required in patents is not greater than that which is reasonable, having regard to the subject matter involved. Minerals Separation, Ltd. v. Hyde, 242 U. S. 261, 37 S. Ct. 82, 61 L. Ed. 286. This well established proposition of law readily lends itself to flexible and practical application in appraising the sufficiency of patent disclosures. That a claim should be clear and definite is, of course, elementary, but the degree of definiteness necessary to satisfy the law varies in accordance with the facts and circumstances of each case. In the case of General Electric Co. v. Wabash Appliance Co., 304 U. S. 364, 58 S. Ct. 899, 901, 82 L. Ed. 1402, the court said:
>
> "' *** Patents, whether basic or for improvements, must comply accurately and precisely with the statutory requirements as to claims of invention or discovery. The limits of a patent must be known for the protection of the patentee, the encouragement of the inventive genius of others, and the assurance that the subject of the patent will be dedicated ultimately to the public. The statute seeks to guard against unreasonable advantages to the patentee and disadvantages to others arising from uncertainty as to their rights. The inventor must 'inform the public during the life of the patent of the limits of the monopoly asserted, so that it may be known which features may be safely used or manufactured without a license, and which may not.' The claims 'measure the invention' ***'
>
> "We cannot conclude that the presumption of correctness attaching to the decision of the Patent Office tribunals in the instant case is sufficient to overcome what we consider to be an aptly and adequately drawn claim, which fully defines the invention and which finds clear and abundant support in the specification of disclosure."

And as was said in S. D. Warren Co. v. Nashua Gummed and Coated Paper Co. (CA 1), 205 F. 2d 602, 605:

> "Particularity of identification and clarity of statement are relative terms when applied to an invention. Some inventions are capable of description in terms of meticulous accuracy, others are not. And this is one of the latter class.-----Reading the claims in the light of the specification we think one skilled in the art could readily apprehend the precise nature of Perry's invention and the limits of its scope. This is all the law requires."

The invention involved is a novel, useful chemical compound. The name of this compound is not known, else it would have been claimed by name, and a patent granted long ago. Applicants have claimed the inventive compound another way, i.e. by recitation of its properties. This method of claiming is not per se objectionable and is often used where the name is not known. There is no requirement that every possible property be listed, only that sufficient properties appear to meet the statutory requirement. As an illustration in another McBee patent 2 849 499, claim 1 reads: "A composition of matter having the empirical formula $C_{10}Cl_{12}$ and melting at 221 to 223 degrees centigrade." (733 O. G. 1054); and in Plaintiff's Exhibit 2, McBee et al U. S. patent 2 516 404, claim 6 reads (JA 106)

> "A compound of the approximate molecular formula $C_{10}H_2O_3SCl_{12}$, having a melting point, in the solid state of about 146 to 147 degrees centigrade, being substantially identical with the product formed by the reaction of chlorosulfonic acid with hexachlorocyclopentadiene and having valuable insecticidal properties."

The main reason for the requirement of 35 USC 112 is so it may be known whether or not a "new" compound infringes the claimed compound. A leading text "The Characterization of Organic Compounds" (JA 111) shows that chemists determine whether or not a "compound encountered in the laboratory is novel, by comparing its properties with those of the known chemical compounds recorded in the chemical literature, and that while this might at first glance, have some of the aspects of the search for the needle in the haystack...the fortunate fact that this great number of organic compounds can be grouped into relatively few classes has been responsible for the development of systematic procedures of identification that make the characterization of a compound, the properties of which are recorded in the literature a comparatively simple matter." Thus by comparison of the properties of an unknown compound with the properties enumerated in the claims, a chemist can tell whether or not the unknown compound is that of the claims.

The claims on appeal list the following properties: color, form, melting point, behavior, solubilities in two types of solvents, and molecular formula. "The Handbook of Chemistry and Physics", Plaintiffs Exhibit 3, (JA 107, 110) shows that all compounds listed are described by their name, formula, molecular weight, crystalline form, color, density (only if a liquid), melting point, boiling point (only if a liquid) and solubilities in various solvents. Except for the name, the claims expressly contain all the information as to the properties regarded as most important by chemists, and even gives an additional characteristic. These compilations of properties in the literature are commonly indexed, not only by name, but by formula and melting point. Thus Plaintiffs Exhibit 3 has a Formula Index (JA 108) wherein all the compounds described therein are grouped by formula, all C_9 compounds in one group, all C_{10} in another group, all C_{11} in another group, etc., and there is a pattern followed within each group, so that in a few seconds it can be ascertained whether any $C_{10}Cl_{10}O$ compound is included. (Mr. Sconce testified that there is none).

This Exhibit also has the customary melting point index (JA 109) wherein for a particular temperature all compounds melting thereat are listed. It will be noted that there is a compound listed which melts at 354 degrees centigrade, but a glance at the compound shows it is totally unlike that claimed.

Workers in the field of organic chlorine compounds, which is that involved here, are fortunate to have the benefit of "Organic Chlorine Compounds", Plaintiffs Exhibit 6, which is a complete compilation of all organic chlorine compounds known as of about 1947. This compilation indexes its compounds in various ways, including by the percentage of chlorine in the compound and by the molecular weight. There are several compounds listed having about 72 percent chlorine, but it is immediately obvious by inspection that none of these has a formula at all close to applicants. Also the highest molecular weight listed is 418, which is much lower than the 493 molecular weight of the claimed compound.

It is thus apparent that the melting point and formula of all known organic compounds, and especially organic chlorine compounds are so classified, that knowledge of these two properties provides an easy check against known compounds. It is true that many compounds may have the same melting point, and many compounds may have the same molecular formulas. However, most organic compounds melt below 300 degrees centigrade. To illustrate, Plaintiffs Exhibit 3 lists the melting points of over 5000 compounds ranging from 200 degrees centigrade to 500 degrees centigrade on some 14 pages (p. 1267 - 1280), but less than 100 melt above 300 degrees centigrade which means that over 98 percent melt below 300 degrees centigrade (see 1267 and 1280 on JA 109). Thus applicants melting point of about 355 degrees centigrade is a very unusual one, and it is not difficult to look up the other characteristics of all known compounds melting within 5 degrees centigrade, either way of that figure. Applicants molecular formula is also unusual. As explained previously, the molecular formula tells a chemist the percentage of each element and also the total molecular weight. The molecular weight 493 is at least 15 percent heavier than any listed in Plaintiffs Exhibit 6 (JA 113). It is possible, albeit unusual, for two compounds to have the same melting point and molecular formula. In this case, the other properties are also compared.

It is submitted to be proper to consider the various properties as somewhat like "fingerprints"; considered separately, any one of these fingerprint identifying characteristics recited by the claims might not exclude all other possible compounds having such selected individual characteristics, but, taken collectively, the recited physical characteristics do absolutely identify one single substance. It is the cumulative effect of the several recited properties which narrows the field to one compound meeting the claims. Perhaps a better analogy is to the Bertillon system for identification of persons by a physical description based upon anthropometric measurements, the cumulative effect of which limits the description to a single individual.

The compound of the present invention is unusual in several respects. It is very hygroscopic, and picks up water from the air, so it is very difficult to be sure whether one has the anhydrous compound, without water, or the hydrated form of the compound, with water. This is the reason why the terms "about" and "approximately" must be used in the claims, since not to do so would mislead the public. If a chemist wrongly assumed that he had the anhydrous compound, then the analysis of the compound would show all or part of a water molecule, H_2O, with the $C_{10}Cl_{10}O$ of the base compound. The compound is also unusual in its composition $C_{10}Cl_{10}O$ and its high molecular weight. Chlorine containing compounds having a very high molecular weight are most rare. The compound is also unusual in that its melting point is so high as to be most rare. The above characteristics are of a type which makes analysis of the compound difficult and subject to a larger variation of accuracy than is permissible in the usual type of organic compound. For this reason, the claims contain the expression "about 355 degrees centigrade" and "approximate molecular formula $C_{10}Cl_{10}O$" since in order to give applicants protection commensurate with the merits of their invention, it is necessary to recognize some permissible latitude in the melting point and molecular formula, and these expressions also serve to warn those who would avoid infringing that they must be cognizant of the higher limits of experimental error present in this type of compound.

Each descriptive characteristic used in the claim has a clear and definite meaning to a chemist. Thus, when a chemist sees the expression "approximate molecular formula $C_{10}Cl_{10}O$," applied to a compound which is hard to obtain in water-free form, and very often has some H_2O present so it is in its hydrated form, then he knows that "approximate" refers only to the possible presence of H_2O.

Thus Finding of Fact 11 is in error that "approximate" could only have significance as indicating that the formula may be varied by a whole number of any one or more of the three kinds of atoms indicated. No variance in the C or Cl would be inferred by a chemist, only in the O and the possible addition of an H. The specification is very definite that exactly 10 chlorine atoms are present (JA 49) and no chemist would doubt that exactly 10 carbon atoms are always present also since it is prepared from $C_{10}H_2Cl_{12}O_3S$. Thus the "approximate" actually modifies the O and the possible presence of H. In any event, the formula under either interpretation is very significant, since Handbooks and other compilations of organic chlorine compounds have formula indices whereby all possible meanings of "approximate molecular formula $C_{10}Cl_{10}O$" may be easily checked. Because of the unusualness of this formula and its approximations, no undue effort would be required.

The melting point expressed as "about 355" has a range of several degrees on either side. Contrary to the Finding of Fact 10, this variance does not mean that the melting point does not have a sufficient "degree of accuracy to make it, by the standards of the organic chemistry art, an identifying characteristic." Note, "The Handbook of Chemistry and Physics" wherein at JA 110, many compounds are shown to have melting points covering a range. A glance at the column headed "Melting Point" shows such melting points as 238-41, 215-20, 220-2, so it is evident, as Mr. Sconce testified, that even with a few degree variation, melting point is significant. Because of the few known compounds melting anywhere near 355 degrees centigrade, and because of the higher margin of error in this excessively high range, any chemist having an unknown compound melting within 5 degrees of 355 would naturally check further before rejecting the possibility that he infringes, so the use of "about" language does not make the claim any more indefinite than it would be without such language.

The other recited characteristics are adequately defined in the specification, which specifies in what way the compound "behaves as a weak organic acid," gives examples of polar and non-polar solvents. These characteristics belong to many compounds, as does the color, and crystal structure, but each of these characteristics is useful as a point of identification to be used in case of doubt, as to the sufficiency of the major characteristics already discussed.

Claim 13, contains all the language of claim 6 plus an expression showing how the compound "is preparable". Thus claim 13 contains an added identifying characteristic. By following the last part of claim 13, an authentic sample of the claimed compound is obtained, and by testing this authentic sample against an unknown compound in a mixed melting point test, a chemist will definitely know whether or not the unknown compound is the same compound as that of the sample. The language "is preparable" is used because such language does not limit the coverage of the claim to compounds prepared only by this method. The specification teaches at least four basic methods of preparing the claimed compound, so it is not meet that the claim be limited to cover the compound only when made by one method. The applicants are entitled to protection commensurate with the scope of their invention, and their invention is the novel compound, no matter how it is made. Should the court feel that claim 6 does not meet the statutory requirement, then certainly claim 13, with its additional, positive, identifying characteristic, does.

CONCLUSION

It is respectfully submitted, in view of the foregoing, that the District Court having reached an erroneous conclusion in this case should be reversed, so that the appellee, Commissioner of Patents, may be instructed to issue Letters Patent based on the application here involved.

Respectfully submitted,

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BRIEF FOR APPELLEE

United States Court of Appeals
For the District of Columbia Circuit

APPEAL NO. 14,704

PURDUE RESEARCH FOUNDATION, *Appellant*

v.

ROBERT C. WATSON, COMMISSIONER OF PATENTS,
Appellee

Appeal from the Judgment of the United States District Court for the District of Columbia

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United States Court of Appeals
For the
District of Columbia Circuit

FILED JAN 5 1959

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STATEMENT OF QUESTIONS PRESENTED

In the opinion of the appellee questions presented by this appeal are:

(1) Whether or not the common ruling of the Patent Office tribunals and the District Court that a claim to a chemical compound is inadequate under 35 U.S.C. 112 is reasonable where the compound is described in terms of its approximate formula and approximate melting point and by reference to the "characteristics," color, crystalline form, solubility, and behavior "as a weak organic acid," which characteristics are shared by many organic compounds, and where the evidence indicates that the compound in fact is not a weak organic acid but a ketone and the "approximate molecular formula" not that attributed to the compound in the application as originally filed?

(2) Whether or not the concurring rulings of the Patent Office examiner, the Patent Office Board of Appeals, and the District Court that a claim is inadequate under 35 U.S.C. 112 are reasonable where the description referred to in question (1) above is supplemented by a reference to the general treatment or process by which the compound is "preparable"?

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United States Court of Appeals
For the District of Columbia Circuit

APPEAL NO. 14,704

PURDUE RESEARCH FOUNDATION, *Appellant*

v.

ROBERT C. WATSON, COMMISSIONER OF PATENTS,
Appellee

Appeal from the Judgment of the United States District Court for the District of Columbia

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BRIEF FOR THE COMMISSIONER OF PATENTS

INTRODUCTION

This is an appeal by Purdue Research Foundation from the judgment (J.A. 140) of the United States District Court for the District of Columbia dismissing its complaint in an action under Section 145, Title 35, of the United States Code, in which action appellant sought to have that court authorize the issuance to it of a patent containing claims 5, 6, 7, 8, and 13 of the application of Earl T. McBee and Jack S. Newcomer, entitled "Composition of Matter," Serial No. 238,186, filed in the United States Patent Office on

July 23, 1951. Purdue Research Foundation is assertedly the assignee of the application.

The Patent Office examiner (J.A. 94, 95), the Patent Office Board of Appeals (J.A. 98-102) and the District Court (J.A. 139 and 140-145) concurred in rejecting the noted claims 5, 6, 7, 8 and 13 (J.A. 61, 62), which are all the claims remaining in the application, on the ground that they do not adequately define the invention in accordance with 35 U.S.C. 112.

COUNTER STATEMENT OF CASE

The McBee and Newcomer application is directed to a chemical compound which is said to be useful in its anhydrous form as a dessicant because of its hygroscopic nature, and as an ingredient in insectidical compositions (J.A. 43 and 44). The application as filed (J.A. 43) stated that this compound was "believed to have the approximate molecular formula $C_{10}HCl_{10}O_2$" and that "the exact elemental composition and structure of the compound are not precisely known at the present time." Over a year after the filing of the application it was amended (J.A. 64 and 65) to change the "approximate molecular formula" of the compound from "$C_{10}HCl_{10}O_2$" to "$C_{10}Cl_{10}O$," which change involved the elimination of one hydrogen atom and one oxygen atom from the formula. Subsequent to the final rejection of the application McBee and Newcomer filed a supplementary oath (J.A. 79-80) which was allegedly applicable to the subject matter of their claims which was disclosed but not claimed in their application as originally filed in the Patent Office.

The specification of the McBee and Newcomer patent application (J.A. 46, 47) states that the composition of the present alleged invention exists in the anhydrous form as well as in the form of various hydrates and that the anhydrous and hydrate forms all give the same melting point in a sealed tube of approximately 355 degrees Centigrade, corrected. A tabulation, in the specification (J.A. 48), of melting point data for "$C_{10}HCl_{10}O_2$" (later amended to read $C_{10}Cl_{10}O$) shows a variation of melting points (degrees Centigrade corrected) from 331 to 350 at "1st Soft;" from 351 to 355 at "1st Meniscus;" and from 351 to 361 at "1st Crystal."

As the District Court's Findings of Fact (J.A. 141, paragraph 5) indicate, counsel for the plaintiff and defendant agreed at trial that claims 5, 7 and 8 of the application in suit should stand or fall with claim 6 and that if the District Court should allow either claim 6 or claim 13, the other claim, claim 13 or claim 6, should not be allowed.

Claims 6 and 13 read as follows:

> 6. A composition of matter which has the following characteristics: it is a white crystalline solid having a sealed tube corrected melting point of about 355 degrees Centigrade, it behaves as a weak organic acid, is very soluble in polar organic solvents, substantially insoluble in non-polar organic solvents, and has the approximate molecular formula $C_{10}Cl_{10}O$.
>
> 13. A composition of matter which has the following characteristics: it is a white crystalline solid having a sealed tube corrected melting point of about 355 degrees centigrade, it behaves as a weak organic acid, is very soluble in polar organic solvents, substantially insoluble in non-polar or-

> ganic solvents and has the approximate formula $C_{10}Cl_{10}O$ and is preparable from a compound having a melting point of approximately 146-147 degrees centigrade, formed by the reaction of hexachlorocylopentadione with chlorosulfonic acid, by treatment with alcoholic alkali and acidification and dehydration.

Claim 5 differs from claim 6 in that it claims the *hydrates* and *salts* of the compound covered by claim 6 as well as the compound itself. (A hydrate has one or more molecules of water added to the basic molecule).

SUMMARY OF ARGUMENT

I. To meet the requirement of particularity and distinctness stated in 35 U.S.C. 112, claims not only "must clearly distinguish what is claimed from what went before" but must also "clearly circumscribe what is foreclosed from future enterprise." The limits of a patent must be known so that it may be ascertained which features may be safely used or manufactured without a license and which may not. Since appellant's claims are directed to a chemical compound, a product which has a fixed molecular composition, these claims must meet a more exacting standard then that applicable to physical processes. By the standards of organic chemistry the characteristics of the claimed chemical compound, as set forth in claims 6 and 13 do not, when taken individually or in combination, adequately and distinctly describe the claimed compound because:

> (a) The melting point range indicated by "about 355°C." is not a significant i~~n~~dentifying element because of its extent.

(b) The "approximate molecular formula" recited in the claims is without identifying significance because the molecular formula claimed was not originally disclosed and because it is qualified in a manner to render the formula meaningless.

(c) The expression "behaves as a weak organic acid" is not only indefinite, as the Board held (J.A. 100), but as the evidence shows is misleading because the compound is in fact not an acid but a ketone.

(d) White color, crystallinity, behavior as a weak organic acid (no matter how interpreted), and solubilities as to polar and nonpolar solvents are shared by many compounds.

II. Reference to the process of preparation of the compound in claim 13 would be justifiable only if the compound could not be otherwise described. If such a manner of description is resorted to the claim must be limited to the product produced by a method identified in the claim. Claim 13, although it refers to a process by which the compound is made, does not limit the intended monopoly by reference to that process, and it is, therefore, not a permissible assertion of monopoly. This claim if patented would have the effect of requiring a prospective manufacturer, seeking to avoid infringement, to carry out the indicated process in order to determine whether his own product, produced by another process, is an infringement.

ARGUMENT

I. Section 112 of Title 35 of the United States Code (quoted, appellant's brief, page 2) states, as follows, the requirement as to an application claim:

> The specification shall conclude with one or more claims particularly pointing out and distinctly claiming the subject matter which the applicant regards as his invention.

This requirement of 35 U.S.C. 112 is essentially that stated in its predecessor, R.S. 4888, 35 U.S.C., 1946 ed., 33, which required an inventor to "particularly point out and distinctly claim the part, improvement, or combination which he claims as his invention or discovery."

The Supreme Court in *United Carbon Company et al* v. *Binney and Smith Company,* 317 U.S. 228, 236, held as follows:

> The statutory requirement of particularity and distinctness in claims is met only when they clearly distinguish what is claimed from what went before in the art and clearly circumscribe what is foreclosed from future enterprise. A zone of uncertainty which enterprise and experimentation may enter only at the risk of infringement of claims would discourage invention only a little less than unequivocal foreclosure of the field. Moreover, the claims must be reasonably clear-cut to enable courts to determine whether novelty and invention are genuine.

In the earlier case of *General Electric Co.* v. *Wabash Appliance Corp.,* 304 U.S. 364, 369, the Supreme Court ruled as follows:

> Patents, whether basic or for improvements, must comply accurately and precisely with the statutory requirement as to claims of invention

> or discovery. The limits of a patent must be known for the protection of the patentee, the encouragement of the inventive genius of others and the assurance that the subject of the patent will be dedicated ultimately to the public. The statute seeks to guard against unreasonable advantages to the patentee and disadvantages of others arising from uncertainty as to their rights. The inventor must—

inform the public during the life of the patent of the limits of the monopoly asserted, so that it may be known which features may be safely used or manufactured without a license and which may not. "

These Supreme Court decisions in cases involving products indicate the general standard to be used to measure the adequacy of the description of an invention in the claims of a patent, but it is believed apparent that the nature of the invention claimed must be taken into account in determining whether the terminology of the claims sufficiently delineates the invention. In the present case the subject matter is a chemical compound and, therefore, necessarily a material with a fixed chemical composition which properly cannot be defined in relative terms of function or purpose. Such relative terms may well suffice, as the decisions quoted by appellant (brief, pages 17, 18, 19) illustrate, in some descriptions of a general process. *Lever Brothers Co.* v. *Procter and Gamble Manufacturing Company* (C.C.A. 4, 1943) 139 F.2d 633, 639; *In re Hudson,* 40 C.C.P.A. 1036, 1040, 205 F.2d 174, 177. Even in claims to a process, however, the discription of a feature of the process in relative terms of effect or purpose may be defective for lack of definiteness or particularity; see, for instance, *Standard Oil Company of California* v. *Tide Water Associated Oil Company* (C.C.A. 3, 1946) 154 F.2d 579, 581; *Standard Brands, Inc.* v. *National Grain Yeast Corporation,* 308 U.S. 34, 38.

The appellant's quotation (brief, page 17) from *Lever Brothers Co.* v. *Procter and Gamble Manufacturing Co. et al.* (C.C.A. 4) 139 F.2d 633, 639, includes a quotation (brief page 17, last paragraph) from the same court's decision in *Procter and Gamble Manufacturing Co.* v. *Refining, Inc.* (C.C.A., 1943) 135 F.2d 900, 906, which replaces by asterisks that part of the latter decision which is most relevant to the present case:

> "This argument, we think, misconceives the nature of the claims in suit. They do not state merely the results to be achieved, but are process claims which set out a series of steps by which the desired ends are attained with sufficient definiteness to inform persons skilled in the art and to limit the extent of the monopoly of the patent."

This Court in *Montgomery* v. *Marzall,* 88 U.S. App. D.C. 281, 284, 189 F.2d 640, 643, not only quoted this portion of the Circuit Court's decision but demonstrated its relevance by the following quoted rulings:

> In like manner, the Patent Office tribunals and the District Court have in this case mistaken the nature of the claims in issue. They are true process claims, in that they describe the acts to be done in manufacturing cast coated paper, including new steps added by appellants to the previously practiced process. The claims are not for the com-

> pounding of ingredients into a new product, nor are they for a manufactured article, the characteristics of which are caused or affected by the materials of which it is made. That is to say, these are not claims for compounding a new coating composition by adding a specified proportion of oil. The coating composition is the same as before.

S. D. Warren Co. v. *Nashua Gummed and Coated Paper Co.* (C.C.A. 1, 1953) 205 F.2d 602, 605, quoted in appellant's brief (page 20), found that the claims to an adhesive composition there involved were not "functional" in the sense of presenting a statement of function at the precise point of novelty. The court (at page 605) noted that the patentee had stressed that there was "no chemical reaction" between the ingredients of this composition, a type of resin and a plasticizer, and (in the portion indicated by asterisks in the appellant's quotation) that the patentee had "described the essence of his invention in detail and with particularity in his specification and listed therein about forty illustrative examples of his film." This decision, therefore, did not deal with a situation where a chemical compound in itself constituted the sole point of novelty.

It is appellee's position that the Findings of Fact of the District Court accurately present the facts of this case and that the District Court's judgment finds ample basis in these Findings. This brief discusses below the individual "characteristics" which appear in the claims. It is emphasized that this treatment of separate characteristics is merely for convenience because, as Finding 16 (J.A. 144) specifies, the recited characteristics do not, when taken *individually or in* *combination,* adequately and distinctly describe the the claimed composition.

(I-a). "Melting Point of About 355 Degrees Centigrade."

The Board's relevant holding (J.A. 99) is as follows:

> The record of this case does not warrant an assumption that the melting point and molecular formula are significant identifying elements. The claim refers to the melting point as "about 355° C." The table on page 6 of the specification gives figures obtained in determining the melting point of several samples of the claimed compound. These figures range from 331 to 351 for the first softening point; 351 to 355 for the meniscus, and 352 to 361 for the first crystals. It appears therefore that the qualifying term "about" is intended to include a variation of up to 5° above or below the figures specified. We do not believe that it can be seriously asserted that a melting point range as wide as indicated defines an identifying characteristic.

The evidence adduced at trial on the melting point supports the Board's holding. Testimony by appellant's witness, Mr. Sconce (J.A. 22), that in his company's laboratory work at Niagara Falls low temperature melting points were determined to an accuracy of a hundredth of one degree but that around 355° C. a "temperature is [of ?] a much wider figure, two degrees, three degrees or four degrees, is quite significant to us," shows that the witness was not attaching significance to melting point ranges as wide as those implicit in the term "about 355° C."

Table 1 of the application in suit (J.A. 48) shows melting points which vary at the "1st crystal" stage of a melting point determination by nine degrees (352° to 361°). [The additional error inherent in the fact that this table does not give the "last crystal point," which Mr. Sconce states (J.A. 21) is used to determine the true melting point, may be ignored because the range of melting points reported would make any error from this omission insignificant by comparison]. Assuming a variation of five degrees (the actual maximum variation from 355°C. being six degrees for "1st crystals"), the possible error could be twice this figure in any comparison with another compound. This arises from the fact that the errors in the determination of melting points could be cumulative, that is, two compounds may be the same and still appear, because of errors in determination, to differ in melting points by ten degrees, or two compounds may have true melting points differing by ten degrees and still appear to be the same compound.

As the quotation from the Board's decision suggests, chemists recognize that melting points must be determined with a high degree of accuracy to be a positive and definite item in the identification of a new compound. Page 2 from the text by McElvain (Plaintiff's Exhibit 4, J.A. 111) which Mr. Sconce referred to in his testimony (J.A. 25) and page 582 from the one volume *Encyclopedia of Chemistry* (Plaintiff's Exhibit 7, J.A. 115) to which counsel for the appellant referred in his opening statement (J.A. 8) both indicate that a melting point is one item of compound identification, but neither suggests that melting points varying within a range as wide as four or five degrees are accurate means of identification.

Mr. Sconce, although he testified (J.A. 22) to the inaccuracy of melting point determinations at high temperatures, did not go as far as to say that such inaccurate high temperature melting points were valid and definite means of description of a new compound. Excerpts from three different works on organic chemistry were received from the Commissioner of Patents at trial to rebut any implication that "quite significant," which the witness applied (J.A. 22) to melting points *correct* to "two degrees, three degrees, or four degrees," means that even such inaccurate melting points are adequate elements of identification. These excerpts were taken from Continental (J.A. 127), English (J.A. 128, 129), and American (J.A. 130) sources to show the recognized standard of accuracy in melting point determination prevailing throughout the field of organic chemistry.

Appellant's brief (pages 24 and 25) points to some melting point data from the "Handbook of Chemistry and Physics" (Plaintiff's Exhibit 3, J.A. 109) in suggesting that "many compounds are shown to have melting points covering a range" and that, "Because of the few known compounds melting anywhere near 355 degrees centrigrade, and because of the higher margin of error in this excessively high range, any chemist having an unknown compound melting within 5 degrees of 355 would naturally check further before rejecting the possibility that he infringes * * *." These suggestions ignore the fact that the table merely presents available "data" on certain compounds [see Explanation of Table, J.A. 107, first paragraph], which were selected to include merely those of "general or commercial importance" and "those commonly met with in the laboratory." The Handbook does not suggest that wide ranges of melting points be accepted as significant identifying information. The Handbook, indeed, identifies each compound [see sample page, J.A. 110] by means of its systematic names and formula.

(I-b). "Approximate Molecular Formula $C_{10}CL_{10}O$"

The Board's decision (J.A. 99) held the "approximate molecular formula $C_{10}Cl_{10}O$" to be without identifying significance in the following language:

> "The molecular formula in the claim refers to the presence of 10 carbon atoms, 10 chlorine atoms and 1 oxygen atom. The original specification stated the formula as also including an additional atom of hydrogen and of oxygen. Since the molecular formula now specified in claim 6 is not derivable from the original specification, it cannot be asserted that the claimed formula constitutes essential identifying information. The statement of the molecular formula is qualified by the term 'approximate'. Qualifying terms of this character are normally employed in claims to compositions of matter and are generally acceptable as providing for insignificant variation from particularly named limiting amounts. The reasoning ordinarily employed in justifying such qualifying terms is inapplicable to a molecular formula since a molecular formula represents a stated number of atoms of each of the kinds indicated and there can be no variation by a fractional portion of an atom. The word 'approximate' used in connection with the formula specified in claim 6 can only have significance as indicating that the

> formula may be varied by a whole number of any one or more of the three kinds of atoms indicated. Such a variation wholly destroys any identifying significance in the formula. Since the molecular formula claimed was not originally disclosed, and since the statement of the molecular formula in the claim is qualified in a manner to render such formula meaningless, we must hold that statement in the claim in this respect is without identifying significance."

Appellant's quandary is apparent from this statement. If the formula recited in claim 6 is of any significance it is matter new to the specification and, as such, prohibited. If the formula recited is a mere approximation it is worthless as a means of identification in a science which, as the organic compound compilation [sample pages, J.A. 108, 110, 112, 113] adduced for appellant demonstrates, identifies its compounds by structures or names indicating these structures.

Appellant's brief (pages 23 and 24) argues that the term "approximate" is present in the claim to allow for the presence of water, H_2O, in the molecule and that "approximate actually modifies the O and possible presence of H." The claim, however, does not so state. If the molecular formula is merely "approximate" there may be, as the Board noted (J.A. 100), a greater variation of any or all elements than the addition of a single oxygen or hydrogen atom. A compound with 9, 10, or 11 carbon atoms, 9, 10, or 11 Chlorine atoms, 1, 2, or no oxygen atoms and an optional hydrogen atom would be covered *if the approximation allowed only a variation of one atom of each* *kind* and no larger variation is excluded by the language of the claim.

If the appellant wanted to allow for water of hydration in its claims, this could be done by merely calling for the hydrate in the claims. The fact that the original claims (J.A. 57, 58) actually *included provision for hydrates* ["(b) hydrates thereof" in claim 1; a "hydrate of the compound defined in Claim 2" in claim 4] and the circumstance that appellant's present claims 5 and 8 (J.A. 61, and 62 with amendment J.A. 65) *expressly include reference to hydrates* ["hydrates of (A)" in claim 5; a "hydrate of the compound defined in claim 6," specified in claim 8] would seem to explode the theory which appellant advances as an excuse for the change in formula and a justification for use of an "approximate formula." There is, moreover, no substitute theory advanced to explain what degree of variation from the formula $C_{10}Cl_{10}O$ is presently intended, or was contemplated in the original application.

Appellant asserts (brief, page 24) that, "No variance in the C or Cl would be inferred by a chemist, only in the O and the possible addition of an H," and that the "specification is very definite that exactly 10 chlorine atoms are present." However, the specification at the page appellant refers to (J.A. 49) states merely that the "presence of ten chlorine atoms in the compound $C_{10}HCl_{10}O_2$ is *indicated* by the analysis * * *" (emphasis supplied). If "no chemist would doubt that 10 carbon atoms are always present" in view of the starting material for appellant's method of preparation (appellant's brief, page 24) the evidence does not demonstrate this to be a fact.

Appellant's suggestion (brief, page 24) that "all possible meanings of 'approximate molecular formula $C_{10}Cl_{10}O$' may easily be checked in handbooks and compilations" like appellant's companion suggestion (brief, page 25) that any chemist would "naturally check further" if he had an "unknown compound melting within five degrees of 355" indicate that appellant believes that its claims may set up a "zone of uncertainty which enterprise and experimentation may enter only at the risk of infringement," in disregard of the ruling in *United Carbon Company et al.* v. *Binney and Smith Company,* 317 U.S. 228, 236. These suggestions also indicate that appellant believes, without apparent legal basis, that a manufacturer's determination of infringement should await the preparation of compilations and formula indices which include appellant's claimed compound.

Appellant's emphasis (brief, pages 22, 23, and 24) on the unusualness of the composition $C_{10}Cl_{10}O$, its unusual molecular weight, and its "rare" high melting point, is in the nature of a prophecy that the handbook (J.A. 107, 110) and "Organic Chlorine Compounds" (J.A. 112, 113), rather than the future development of the art, will provide an indication of the difficulty which will be encountered in the future in distinguishing the claimed compound from possible infringements. Such a prophecy on the basis of a compilation of certain compounds of general or commercial importance (J.A. 107, first paragraph) and a collection of organic chlorine compound dated 1948 (J.A. 22) would appear not only ill based but irrelevant to the issue.

(I-c). "Behaves as a Weak Organic Acid"

The Board's decision held (J.A. 100) that the expression "behaves as a weak organic acid," like the general statement of solubility in the claims, is so broad as to obviously include many compounds of entirely different characteristics. The Board stated, "there is no assertion the compound is in fact an organic acid and the statement in question is indefinite."

The testimony of appellant's witness at trial showed that the Board's statement was conservative. Appellant's witness testified (J.A. 24, 25) that it is now fairly well accepted that the claimed compound is in fact a *ketone* but that when it reacts with water it 'puts hydrogen ions in solution" which "can be titrated * * * just as any weak organic acid." The witness asserted (J.A. 25) that the claimed compound has another characteristic of weak organic acids in that it will "react with amines and other basic compounds."

The significance of this testimony lies in the fact (which was not disclosed in the original application as filed) that the compound, which is said to "behave as a weak organic acid," is in fact a ketone. If this compound behaved in every respect as an organic acid it would not be a ketone but an acid so that the compound must be lacking in at least one important and essential characteristic of an acid. The quoted expression is inaccurate if it is intended to mean that the claimed compound is a weak organic acid; and it is indefinite and of uncertain scope if it is intended to mean that the compound is not an acid but has some of the characteristics of an acid *which are unspecified* in the claim and lacks other characteristics *which are* also *unspecified.*

The circumstance that the compound is a ketone, although not so identified in appellant's application, makes relevant the quotation in appellant's brief (pages 20 and 21) from a "leading text" (J.A. 111) to the effect that the grouping of compounds into relatively few classes has made the characterization of a compound a comparatively simple matter. Obviously without an accurate indication of the compound's "class" this simplicity would be lacking.

(I-d). General Characteristics

The appellant's statement (brief, page 25, first complete paragraph) that "these characteristics [referring to the solubility characteristics, and behavior as a weak acid] belong to many compounds, as does the color, and crystalline structure" agrees with the Board's holding (J.A. 100, first complete paragraph) as to these general characteristics. There is no reason to believe that such general characteristics (even assuming that behavior as a weak organic acid is a definite characteristic) definitely identify the claimed compound when the molecular weight and melting point are uncertain.

II. It is well accepted that claims for a product which is defined by the process of producing it may be allowed only where the product involves invention and cannot be defined except by the process used in its creation. *In re Brown,* 58 App. D.C. 285, 29 F. (2d) 873; *In re Moeller,* 28 C.C.P.A. 932, 934, 117 F. 2d 565. 567. Where the product is so described in terms of its process of production, however, the protection is limited to the products of the process referred to in the claim. *In re Moeller, supra,* at 28 C.C.P.A. 936, 117 F. 2d 936; *General Electric Co.* v.

Wabash Appliance Corp. et al., 304 U.S. 364, 373, *Cochrane* v. *Badische Aniline and Soda Fabrik.*, 111 U.S. 293, 310.

Claim 13 adds to the recited "characteristics" of claim 6 the process description "preparable from a compound * * * by treatment with alcoholic alkali and acidification and dehydration."

This process reference, as appellant admits (brief, page 25, final paragraph), would not limit appellant's monopoly to the product of the process referred to in claim 13 but claim 13 would cover the compound no matter by what process produced. Claim 13, according to the ruling of *In re Moeller, supra,* and the noted decisions of the Supreme Court on which that ruling is based, is not a proper assertion of the bounds of appellant's intended monopoly. Were such a mode of claiming permissible infringement of claim 13 could not be determined by a prospective manufacturer of a new chemical product *without carrying out the general process* referred to in the claim and comparing any product obtained with his own chemical product. Then, the appellant suggests (brief, page 25), "by testing this authentic sample against an unknown compound in a mixed melting point test, a chemist will definitely know whether or not the unknown compound is the same compound as that of the sample."

This suggestion that the prospective manufacturer, or chemist, must, to avoid all possibility of infringement, carry out the appellant's generally indicated process in claim 13 to obtain a basis for comparison would seem indeed an amazing conception of the purpose to be served by a claim in stating the exact extent of an asserted monopoly and clearly is inconsistent with the principle that no inventor may compel independent experimentation by others to ascertain the bounds of his claims. *Standard Oil Company of California* v. *Tide Water Associated Oil Company* (C.C.A. 3, 1946) 154 F. 2d 579, 582; *Standard Brands, Inc.* v. *National Grain Yeast Corp.* (C.C.A. 4) 101 F. 2d 814, 822, *affirmed* 308 U.S. 34.

Since, as a practical matter, a claim must be sufficiently definite to allow a Patent Office examiner to determine, without experimentation, whether a product is novel and inventive, the presentation of claims of the type of claim 13 would make effective examination of such claims a practical impossibility and place an unreasonable burden on science and the practical arts.

In view of the indefinite melting point of the claimed compound set forth in appellant's specification (J.A. 48, Table I) and the fact that a melting point determination is "complicated also by the purity of the original compound" (J.A. 26, testimony beginning line 8 from bottom of page), it may reasonably be doubted whether the appellant's suggestion (brief, page 25) that, after the preparation of an "authentic" sample by the process indicated in claim 13, this sample may be compared by a "mixed melting point test" with an unknown to positively establish the identity or lack of identity of the two compounds is well based. As the Board held (J.A. 101), to permit claims in broad enough language to include products of similar character produced by other processes would in general require workers in the art to not only carry out the process indicated in the claims but also *to determine the identity of the product where the inventor himself has been unable or unwilling to do so.*

Appellant's brief (page 20) quotes two patent claims, apparently to suggest that these claims provide some sort of standard of claim definiteness. The reference to the first, assertedly appearing in patent No. 2,849,499, is improper since it is not a part of the record of this case. The second claim, that of patent No. 2,516,404 (J.A. 103), is apparently offered to show the previous allowance of a product claim, including a process reference, which does not clearly limit its asserted monopoly to the product of the indicated process. The validity of this claim was not, however, determined by the Board of Appeals. The Board's discussion of this type of claim (J.A. 101) indicates that only in an exceptional situation where the balance of a claim substantially identifies and defines the novel composition and the added statement concerning process are ^un necessary from the standpoint of such identification would it be proper to permit process references in the claim. It is unnecessary for this Court to determine whether this exception is valid in view of *In re Moeller, supra,* and *In re Brown, supra,* because if the "characteristics" recited in claim 13 are insufficient to describe the invention the exception would have no present application.

CONCLUSION

It is respectfully submitted that each of the claims in suit fails to describe the subject compound with the distinctions and particularity required to define the limits of the asserted monopoly and to comply with the requirements of 35 U.S.C. 112; that the evidence meets the applicable test recognized by *Koebel et al.* v. *Coe;* 70 App. D.C. 261, 105 F. 2d 784, in that it, at the least, establishes that the claims in issue "may reasonably be thought indefinite, so as to sustain the presumption that the Patent Office and the district court were right in denying a patent;" and that the judgment of the District Court should be affirmed.

Respectfully submitted,

CLARENCE W. MOORE,
Solicitor,
United States Patent Office,

ARTHUR H. BEHRENS
Of Counsel.

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REPLY BRIEF FOR APPELLANT

United States Court of Appeals

FOR THE DISTRICT OF COLUMBIA CIRCUIT

United States Court of Appeals
For the
District of Columbia Circuit
FILED JAN 19 1959
Joseph W. Stewart
CLERK

No. 14,704

PURDUE RESEARCH FOUNDATION,

Appellant,

v.

ROBERT C. WATSON,
Commissioner of Patents,

Appellee.

APPEAL FROM THE JUDGMENT OF THE UNITED STATES DISTRICT COURT FOR THE DISTRICT OF COLUMBIA

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Washington, D. C. ROBERT I. THIEL Printer EX 3-0625

UNITED STATES COURT OF APPEALS
For The District Of Columbia Circuit

No. 14,704

PURDUE RESEARCH FOUNDATION,

Appellant,

v.

ROBERT C. WATSON,
Commissioner of Patents,

Appellee.

APPEAL FROM THE JUDGMENT OF THE UNITED STATES
DISTRICT COURT FOR THE DISTRICT OF COLUMBIA

REPLY BRIEF FOR APPELLANT

Appellee's brief relies heavily on two Supreme Court cases, and the purpose of this reply brief is to distinguish the fact situations in those cases from that of the instant case.

In United Carbon Company et al. v. Binney and Smith Company, 317 U.S. 228, 63 S.Ct. 165, a typical claim read as follows:

> "1. Substantially pure carbon black in the form of
> commercially uniform, comparatively small,
> rounded, smooth aggregates having a spongy or
> porous interior."

It can be seen that the claim was directed to a particular form of carbon black. Carbon black itself was old, having been manufactured for some 50 years, and thus the problem before the Court was whether or not the claim distinguished from the known carbon black of the art. The Court rightly held that such terms as "commercially uniform", "comparatively small", "rounded", "smooth", "spongy" and "porous" had meaning only in a functional sense i. e. if a particular carbon black worked in actual processes wherein this claimed type of carbon black was used, then it would infringe the claim, and that "..... the claims are but inaccurate suggestions of the functions of the product, and fall afoul of the rule that a patentee may not broaden his claims by describing the product in terms of function", (317 U. S. at 234). And as the Court also said "... in the view we take of the claims in suit, a mere reading of prior art patents shows how, if they are read with the liberality and inclusiveness claimed for those in suit, they describe products, if not identical, at least of confusing similarity." (317 U. S. at 237). Thus the ground upon which the claim was held invalid was not for lack of "definiteness" but rather for being functional and for being so broad as to read on the prior art because of indefiniteness.

In General Electric Co. v. Wabash Appliance Corp., 304 U. S. 364, 58 S. Ct. 899, a typical claim read as follows:

> "25. A filament for electric incandescent lamps or other devices, composed substantially of tungsten and made up mainly of a number of comparatively large grains of such size and contour as to prevent substantial sagging and offsetting during a normal or commercially useful life for such a lamp or other device."

The claim was directed to a particular type of tungsten filament. Tungsten filaments were old, and the Court held that the claim language just did not distinguish from what was old but in fact "serves aptly to describe the product of earlier manufacture" (304 U. S. at 370). Further the court held "The claim uses indeterminate adjectives which describe the function of the grains to the exclusion of any structural definition, and thus falls within the condemnation of the doctrine that a patentee may not broaden his product claims by describing the product in terms of function." (304 U.S. at 371)

Thus in this case, as in the case of United Carbon et al. v. Binney and Smith Company, supra, the products claimed were old per se and the novelty was only in the particular form of the product. The real reasons why the claims were held invalid resided in their being so broad as to include prior art materials and because the purported limitations were expressed only in functional language.

In the instant case, there is no allegation that appellant's claims are functional or that they read on prior art. Appellant's claimed compound is new, and not just a new form of an old compound. The major identifying characteristics used by appellant are not functional use characteristics, but are numerically stated, and the expressions "about" and "approximately" qualify precise numerical quantities in the same fashion that other similar words of art, such as "substantially", are commonly used in patent law to qualify precise limitations, so as not to cheat the inventor of the fruits of his contribution.

In view of the above, it seems clear that the proper method of construing appellant's claims is as stated in the cases cited in the argument of the main brief and not by the harsher rule which appellee asks for on the basis of cases which are not here applicable.

All of appellee's other arguments are believed adequately answered in appellant's main brief.

Respectfully submitted,
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Attorney for Appellant.

Of Counsel:

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United States Court of Appeals
For the
District of Columbia Circuit

FILED NOV 24 1958

Joseph W. Stewart
CLERK

JOINT APPENDIX

United States Court of Appeals

FOR THE DISTRICT OF COLUMBIA CIRCUIT

No. 14,704

PURDUE RESEARCH FOUNDATION,

Appellant,

v.

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Commissioner,
U. S. Patent Office,

Appellee.

APPEAL FROM THE UNITED STATES DISTRICT COURT
FOR THE DISTRICT OF COLUMBIA

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UNITED STATES COURT OF APPEALS

For The District Of Columbia Circuit

No. 14,704

PURDUE RESEARCH FOUNDATION,

Appellant

v.

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U. S. Patent Office,

Appellee.

APPEAL FROM THE UNITED STATES DISTRICT COURT
FOR THE DISTRICT OF COLUMBIA

JOINT APPENDIX

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JOINT APPENDIX

UNITED STATES DISTRICT COURT
FOR THE DISTRICT OF COLUMBIA

[Filed, February 17, 1956]

1956 February 16
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PRF 4.1-517

PURDUE RESEARCH FOUNDATION :
Lafayette, Indiana :
Plaintiff :
v. :
ROBERT C. WATSON, :
Commissioner of Patents, :
Defendant :

CIVIL ACTION 714-'56

COMPLAINT

(Letters Patent)

To the Honorable Judges of the District Court
for the District of Columbia:

1. This action arises under United States Code Title 35, Section 145. No appeal has been filed in The United States Court of Customs and Patent Appeals.

2. Plaintiff, Purdue Research Foundation, is an Indiana Corporation, having a principal place of business at Lafayette, Indiana.

3. Defendant, Robert C. Watson, is a citizen of the United States, is Commissioner of Patents, and as such a legal resident of the District of Columbia, and is sued herein in his official capacity as Commissioner of Patents.

4. On 1951 July 23, Earl T. McBee, 420 Forest Hill Drive, West Lafayette, Indiana and Jack S. Newcomer, of Hooker Electrochemical Company, Niagara Falls, New York, filed in the United States Patent Office an application For Letters Patent Serial 238 186, entitled "COMPOSITION OF MATTER".

5. Said application for Letters Patent was by an unrecorded instrument in writing, executed 1951 July 6, assigned and transferred to Purdue Research Foundation.

6. Claims 5, 6, 7, 8, and 13 were finally rejected by the Primary Examiner and an appeal was taken to the Patent Office Board of Appeals, which tribunal, by its decision 1955 December 19, affirmed the action of the Primary Examiner as to all of the aforesaid claim and, by this action, Defendant refused and still refuses to grant a patent on said application containing said Claims 5, 6, 7, 8 and 13, inclusive.

The aforesaid claims which are here in issue are as follows:

> 5. A material selected from the group consisting of (A) a composition of matter which has the following characteristics: it is a white crystalline solid having a sealed tube corrected melting point of about 355 degrees Centigrade, it behaves as a weak organic acid, is very soluble in polar organic solvents, is substantially insoluble in non-polar organic solvents, and has the approximate molecular formula $C_{10}Cl_{10}O$; (B) hydrates of (A); and, (C) salts of (A).
>
> 6. A composition of matter which has the following characteristics; it is a white crystalline solide having a sealed tube corrected melting point of about 355 degrees Centigrade, it behaves as a weak organic acid, is very soluble in polar organic solvents, substantially insoluble in non-polar organic solvents, and has the approximate molecular formula $C_{10}Cl_{10}O$.
>
> 7. A salt of the compound defined in Claim 6.
>
> 8. A hydrate of the compound defined in Claim 6.

13. A composition of matter which has the following characteristics: it is a white crystalline solide having a sealed tube corrected melting point of about 355 degrees Centigrade, it behaves as a weak organic acid, is very soluble in polar organic solvents, substantially insoluble in non-polar organic solvents and has the approximate formula $C_{10}Cl_{10}O$ and is preparable from a compound having a melting point of approximately 146-147 degrees Centigrade, formed by the reaction of hexachlorocyclopentadiene with chlorosulfonic acid, by treatment with alcoholic alkali and acidification and dehydration.

7. Plaintiff hereby makes profert of a certified copy of the aforesaid application for Letters Patent, Serial 238 186, and all proceedings and papers in the file thereof, said copy to be available as and when this Honorable Court shall direct.

8. The above-recited Claims 5, 6, 7, 8 and 13, of the aforesaid application are considered to be patentable by the plaintiff.

9. Plaintiff asserts it was error in fact and in law on the part of the Defendant, by the Primary Examiner, and the Patent Office Board of Appeals to refuse said claims of the application.

10. Plaintiff asserts that it is legally entitled to receive a patent for the invention disclosed in said application and claimed in said claims.

WHEREFORE, Plaintiff respectfully prays as follows:

A. For a decree that Plaintiff be entitled to receive Letters Patent on the subject matter contained in the aforesaid application.

B. For a decree pursuant to United States Code Title 35 Section 145, authorizing the Commissioner of Patents to issue Letters Patent on said "COMPOSITION OF MATTER", in due form of law and as prescribed by the statutes.

C. That Plaintiff have such other and further relief as the nature of the case shall admit or require and as may be just and equitable.

Respectfully,

/s/ James B. VanderKelen
Attorney for Plaintiff

* * *

[Filed, April 5, 1956]

ANSWER TO THE COMPLAINT

To the Honorable the Judges of the United States District Court for the District of Columbia.

1, 2, 3, 4. The defendant admits the allegations of paragraphs 1, 2, 3, and 4 of the complaint.

5. The defendant states that he is without knowledge or information sufficient to form a belief as to the truth of the allegations of paragraph 5 of the complaint.

6, 7. He admits the allegations of paragraphs 6 and 7 of the complaint.

8. The defendant admits that claims 5, 6, 7, 8, and 13 of application Serial No. 238,186 are considered to be patentable by the plaintiff, but he denies that they are patentable.

9, 10. The defendant denies the allegation of paragraphs 9 and 10 of the complaint.

FURTHER ANSWERING, the defendant states that he properly has refused to grant a patent on the application here involved containing any of the claims set forth in the complaint for the reasons given in the Examiner's answer and in the decision of the Board of Appeals in said application. Profert of copies of said answer and decision is hereby made.

Respectfully submitted,

/s/ C. W. Moore
Solicitor, U.S. Patent Office,
Attorney for Defendant.

April 4, 1956

[Certificate of Service]

TRANSCRIPT OF PROCEEDINGS

Washington, D. C.,
January 24, 1958.

The above-entitled action came on for trial on the merits, before the HONORABLE JOSEPH C. McGARRAGHY, United States District Judge, at 1:45 o'clock p. m.

APPEARANCES:

On behalf of the Plaintiff:

HERBERT I. SHERMAN, ESQ.
DEAN LAURENCE, ESQ.
PETER F. CASELLA, ESQ.
MRS. MARGARET LAURENCE

On behalf of the Defendant:

ARTHUR H. BEHRENS, ESQ.

- - -

PROCEEDINGS

DEPUTY CLERK: Purdue Research Foundation v. Watson.

THE COURT: Gentlemen, are you ready to proceed?

MR. SHERMAN: Your Honor, I should like to move the admission for the purposes of this case only of Mr. Dean Laurence, member of the bar of the United States Supreme Court, and he will conduct the case.

THE COURT: Very happy to admit you for the purposes of this case.

MR. LAURENCE: Thank you, Your Honor. May it please the Court, this is a patent case, a suit against the Commissioner to compel the issuance of a patent.

You know, in patent cases, I always feel rather odd when they come on right after one of those human interest cases like you just finished, and it is quite often rather dry, I feel.

This case is relatively simple. The subject matter is chemical and it is complicated only, I feel, in the chemical fact side. There is no prior art. We have certain claims which we ask the Patent Office to allow in an application assigned to the Purdue Research

Foundation, which is in Lafayette, Indiana, the plaintiff here; it is a non-profit foundation associated with Purdue University.

The applicant in the patent application before the Patent Office was Dr. Earl T. McBee. Dr. McBee is the head of the chemistry de-
5 partment of Purdue University, which is the largest chemistry department in the world, over 7,000 students taking chemistry there. He has with him Jack S. Newcomer, who is also a Ph.D., obtained his doctorate degree under Dr. McBee as a graduate fellow. They were co-inventors of this subject matter.

Now, there is just one question. As I mentioned, there is no art. The subject matter is patentable, if we have defined it properly.

THE COURT: I was going to ask you a question, which probably will indicate how little I know about it, can you tell me what the subject matter is?

MR. LAURENCE: Yes, I will endeavor to do that right here. It is a chemical compound. If I may inquire, has the Court had any chemistry or you have been educated at least in some chemistry cases?

THE COURT: I have had some chemistry cases.

MR. LAURENCE: All right. A chemical compound is a type of composition of matter. Compositions of matter are one of the statutory classes of patentable subject matter and there are two kinds of compositions of matter, one is made up of mixtures such as sugar and salt and if I put those two together, I have a composition of matter and it might do something. This is not that type. The other type is the so-called true chemical compound. Now, those are made up of a combina-
6 tion of chemical elements, the atoms of elements, arranged in a particular fashion and they arrange themselves in what are known as molecules and each molecule is just like every other molecule and they have unvarying proportions of the atoms within each molecule.

You see, in the former so-called mixtures, you could have ten per cent of sugar and ninety per cent of starch, or whatever it might be, and you could vary proportions all over. Now, when you get into chemical compounds, then the atoms in there are always in exactly the same proportion in each molecule. A molecule made in Germany is just like a molecule made here or in Japan or any other place.

Now, in this case, the composition, I will go to the last criterion we have for it, has the approximate molecular formula $C_{10}Cl_{10}O$. Here is what that means. It means that each one of these molecules, that I talk about being the same no matter where you may be in the world, has ten carbon atoms in it, and each one of those molecules has ten chlorine atoms in it and one oxygen atom.

Now, our trouble is simple. If we knew exactly what the molecular configuration was, just how these atoms were arranged in this molecule, we wouldn't be in this courtroom because the subject matter is otherwise patentable. If you can define your chemical compound by name, you have no problem. The Patent Office allows dozens of patents
7 every Tuesday on compounds which are defined by name and otherwise patentable. But we don't know to this day the structure, there have been a lot of papers published on the structure of this thing and we still don't know the exact structure.

Now, that doesn't mean that we are not entitled to a patent, in our view, because patents are issued where you do not know the exact structure; it is not requisite that you know it, any more than it is requisite that you know the method by which a chemical reaction proceeds or the principle upon which a particular mechanism operates. You don't need to know that, all you need to tell the public is, look, here is a set of characteristics for this substance we have and that set of characteristics can define only one thing. It is a description of a piece of real estate which starts at this bench mark and goes around the property and back to here, and it can't define any other compound, it can only define this specific compound. And that is adequate because then a patent lawyer can advise the client, well, you do or you don't infringe, subsequently, and it is easy to ascertain whether something which might be made by some other party comes within the scope of the claim.

Now, our claim reads like this. It calls for a composition of matter which has certain characteristics. Now, as I mentioned, if we

knew exactly what the structure was, how these ten atoms of carbon
8 and chlorine and the atom of oxygen were arranged in a molecule, I would simply say the name. In accordance with accepted chemical nomenclature, I would say the name right here and that would be the end of the claim and we would have the claim and go away, it would be allowable here. But since I can't tell you, although the applicant has worked on this thing for years and, incidentally, a licensee which is The Hooker Electrochemical Company of Niagara Falls, I think it is the fourth or fifth largest chemical company, also has had their people work on the thing and I have a witness, Mr. Sconce, here from the company as an expert today in this matter, they can't tell us what the structure is yet.

So I have to recite characteristics which define the compound. First of all, we say it is a white crystalline solid. Now, you say, well, that doesn't mean very much; yes, it does. First of all, it is a solid and not a liquid and not a gas. This is by a process of elimination, I have eliminated a lot of things by doing that; it is crystalline and a white crystalline solid.

Then I have given the melting point, a sealed tube corrected melting point of about 355 degrees Centigrade. Now, the melting point is a very important characteristic, I might say, in fingerprinting an organic compound. For example, I have here the Encyclopedia of Chemistry by Clark and Hawley, and at page 582, there is this sentence:

9 > "Melting point determinations have been particularly useful to the organic chemist for identification and characterization of compounds and as a criterion of purity."

Skipping to another sentence:

> "The melting point can be used to identify an unknown compound."

This was published in 1957 by Reinhold Publishing Company.

Now, I will go on to some other properties. By the way, I might say the word "about" occurs in here before the melting point and the Court may wonder why we didn't say exactly a certain melting point.

It is not too easy to say exactly what the melting point is when you get in the higher ranges, and 335 degrees Centigrade is pretty hot. To give the Court some idea, lead melts at 327 degrees Centigrade, so this is above the temperature of molten lead. Your ability to determine exactly the melting point of a chemical compound varies with the range in which you are endeavoring to determine it. If you are down a little above room temperature, you can determine it down to a hundredth of a degree, but when you get up in this very high range, then you can't determine it exactly. So the limitation "about 335 degrees Centigrade", we think is justified here.

Now, we have a second criterion and that is the compound "behaves as a weak organic acid." The Board of Appeals in the Patent Office, I think mistakenly assumed that we were saying it was a weak organic acid. We didn't say that, we said the compound behaves as a weak organic acid. Now, that is a fingerprint for determining what we have. For example, the term "weak organic acid," they say is indefinite, apart from assuming that they said we had one, but it isn't, because the term "organic acid," of course is well known to chemists. And so what we are quarreling about is the word "weak," whether we should say weak or strong. Now, there are two kinds of acids. A chemist generally considers acids which are like muriatic acid, you know, battery acid, the chemical name is hydrochloric acid or sulfuric acid, nitric acid, those types of acids are called strong acids and chemists understand generally what is meant by it.

Now, weak acids are things like vinegar, in the acetic acid of vinegar, and the term weak or strong simply relates to what is called the dissociation constant. It is a term used by chemists to tell us how much hydrogen ion has been liberated in the water solution, so that you know whether it is relatively strong or relatively weak; that is all chemists are after, generally. And in this same Encyclopedia to which I previously made reference, at page 11 there is a note on acids: "The dissociation constant has a larger value for strong acids than for weak ones." The book itself is no more definite than that.

11 THE COURT: What do you mean by the word "behaves"?

MR. LAURENCE: By behaves, what we mean is this, I will explain it and the expert will later, it means that when you put an organic acid, let's take some acetic acid and put it in water, now we have made vinegar, the acetic acid dissociates and liberates some free hydrogen ion in the aqueous solution. Now, we mean that if we take our compound and we put it in water, that it will dissociate and give us a free hydrogen ion in the water just like a weak organic acid does. We don't know that it is an acid and we very much doubt that it is actually, per se, an acid it behaves like one when you put it in water.

Now, the next characteristic which we recite in here is this, we say that this substance "is very soluble in polar organic solvents." Now, that is another fingerprint. What we say is that if you have a beaker or test tube of a polar organic solvent, and the term is well known to chemists, if I said to any chemist "polar organic solvent, get me a bottleful of that," he would go and get one, there are plenty of them known. I say it is very soluble, in that he would put some in and try it and he would find it dissolves very readily.

Now, it is "substantially insoluble in non-polar organic solvents." That is another fingerprint.

And then we come to the last fingerprint, which is this, that the approximate molecular formula is $C_{10}C1_{10}0$.

12 Now, I want to point out that if we take any standard handbook, Lange's Handbook of Chemistry or the Handbook of Chemistry and Physics by the Chemical Rubber Publishing Company, in each book we will find some seven or eight thousand organic compounds listed and there are page after page here in the book of physical constants of organic compounds. Now, these are the way in which chemists identify compounds and if we look at them, it goes like this, we have a name -- well, here, unfortunately, we can't give a name so we lack that characteristic. Next, you have a formula and the formulas are set up in precisely the same form that I have here, $C_{10}C1_{10}0$. Any of these, if I were to read one, $C_5H_{11}3N$, $Br_3C_6H_3$, that simply means a recitation

of what atoms are in there and how many of each atom. That is a standard way of doing it.

Then they give the formula weight. We didn't give it in the claim, but all you have to do is calculate it, it is known that the atomic weight of carbon is 12, so you would multiply 12 times 10, chlorine is 35 1/2, you would multiply 10 times 35 1/2, and oxygen is 16. Everyone knows that, there will be no quarrel about that so I didn't have to give that, it is implicit in the claim.

The next column heading is "crystalline form and color." All right, we gave it, white crystalline solid.

The next item is specific gravity, which you do not ordinarily use with a solid, that is for liquids.

13 The next item is melting point in degrees Centigrade, we gave it.

The next item is boiling point in degrees Centigrade. We don't give that because it is a solid; that again, in the case of compounds, is a liquid.

Then solubility in three different types of solvents. Now, one of the solvents is water, another is alcohol, another is ether. We could have recited it, I believe that the specification does give the solubility in some ten or fifteen different solvents, it tell us what it is. And remember this, that in patent law we have the doctrine that the claim, while it must be definite, nevertheless the dictionary for the claim language is the specification, it is a shorthand form of what is set forth in the specification. So that if you wanted to know which compounds are polar organic solvents, which are non-polar, and in which ones the substance was tested to determine whether it was very soluble or substantially insoluble, you would go back to the specification and do it.

Now, what we say in substance is this, that if you take any one of these characteristics, it would not necessarily define with the particularity required by the statute, 35 U.S.C. 112, the compound. That is true, it might not; although many patents have been granted where you have nothing more than the melting point of the compound and its empirical molecular formula. But we say if you take the cumulative

14 effect of all of these fingerprint characteristics, you can't get anything but this one compound.

Now, we have one claim -- I may say that counsel for the Solicitor's Office and I have pretty much agreed upon this view of the case. There are, I believe, five claims on appeal. May I offer the Court Exhibit 1 of Purdue Research Foundation, the file history of this application in the Patent Office. The Court will find at page 63, the claims which are on appeal.

DEPUTY CLERK: Plaintiff's Exhibit No. 1.

THE COURT: It will be received.

(Certified copy of file wrapper of Application No. 238,186, was marked Plaintiff's Exhibit No. 1 for identification and received in evidence.)

MR. LAURENCE: Now, claim 6 is the one on the blackboard. Counsel and I have agreed that if the Court finds claim 6 allowable, then claims 5, 7 and 8 go along with it and, conversely, if the Court finds claim 6 not allowable, then claims 5, 7 and 8 are not allowable.

Now, we come to claim 13, which differs from claim 6 in this fashion. It is precisely like claim 6, but it adds another fingerprint characteristic and that is we set forth how you can make this compound so that if, by any chance, some day a compound popped up that had all of these identifying characteristics and you still weren't sure, you would inquire, well, how did the man make it? So we added a method

15 of making the compound on as another identifying characteristic. Now, we did add it in this way. We said, "is preparable from a compound having" a certain melting point. Incidentally, that compound at the bottom of claim 13, the last four lines, is preparable from a compound having a certain melting point and formed in a certain way, is the subject matter of United States patent 2,516,404, granted to McBee and Newcomer, the applicants here, and I will offer in evidence as Plaintiff's 2, a copy of this patent, and the compound is defined as claim 7 in that patent already patented to us. In other words, we already have a patent on the intermediate compound from which you make the one that we claim here.

THE COURT: It will be received.

(United States Patent No. 2, 516, 404, was
marked Plaintiff's Exhibit No. 2 for
identification and received in evidence.)

MR. LAURENCE: Now, counsel for the Solicitor's Office and I feel that if the Court should allow Claim 6, then claim 13 ought not to be allowed, because there is a sort of unwritten rule in the Patent Office that if you get the allowance of a claim without anything said about how it is made, you aren't entitled to have a claim saying how it is made, and vice-versa. The Court could allow claim 13 and not claim 6, but probably both ought not to be allowed.

16 I think we could go now to a witness, taking up for a moment the things which the Board of Appeals set forth with which we take issue. I may say that our main contention, apart from the fact that we feel that these fingerprints are enough, is that the Board of Appeals erred in some of its fact statements, findings of fact, you might call them, they aren't set forth quite that way, in its opinion.

Does the Court have any questions on the chemistry?

THE COURT: No, you may proceed.

MR. LAURENCE: I will call Mr. Sconce.

THE COURT: Are you planning to file briefs in this case?

MR. LAURENCE: What is the pleasure of the Court?

THE COURT: I suspect briefs might be helpful to me.

MR. LAURENCE: Very happy to do so. I may say that I don't believe we will be over, perhaps, three-quarters of an hour.

THE COURT: Take as much time as you like; we can sit late, if we have to.

MR. BEHRENS: Should not defendant's counsel give an opening statement?

THE COURT: I think perhaps so.

MR. LAURENCE: Very well.

MR. BEHRENS: I regret, Your Honor, that I have to open up by disagreeing with counsel for the plaintiff on a very simple and probably
17 irrelevant matter and that is the simplicity of this case.

THE COURT: I haven't found any of them very simple.

MR. BEHRENS: That has been my experience, Your Honor. This case is maybe particularly difficult for a Judge or for the Court because, by the law of this Circuit, the question of whether or not a claim is adequate to define the invention in an application is a matter of law for the Judge and not a matter to be determined by the testimony of witnesses. The Patent Office is not, however, contending that the witness' testimony cannot be adduced to help in instruction of the Court in the matter.

This is an action brought under 35 U.S.C. 145 and it directly involves a section of the statute, 35 U.S.C. 112, which requires that a claim in an application particularly point out and distinctly claim the subject matter which the applicant regards as his invention.

It was the position of the Board in this case that this claim, claim 6, which may be taken as representative of one group of claims in the case, and claim 13, the other segment of the claim, do not particularly point out and distinctly claim this subject matter. The statement of the Board of Appeals in this case is a direct and I believe clear statement of the position of the Patent Office in this trial; I would hesitate to improve very much on it, but I would like to comment that the Board
18 of Appeals considered all these factors, all these attempted means of identification in this case, and concluded, and the decision of the case indicates it was a careful conclusion, that the statement of all these items is not sufficient to identify a compound to be given the protection of a United States patent.

This matter of the white crystalline solid, of course, is not very helpful. I think within our common experience we can think of numerous crystalline solids, salt, for example, sugar, no doubt, and other compounds which are white and crystalline and solid.

Now, included in this first statement is a reference to a corrected melting point. I think for the purposes of this case, we can forget about the "corrected", we will call it melting point of about 355 degrees Centigrade. Now, the Board commented with respect to this melting point, perhaps the statement of the Board itself would be the best way of putting this:

> "The record of this case does not warrant an assumption that the melting point and molecular formula are significant identifying elements."

And the Board stated:

> "The table on page 6 of the specification gives figures obtained in determining the melting point of several samples of the claimed compound. These figures range
> 19 from 331 to 351 for the first softening point; 351 to 355 for the meniscus, and 352 to 361 for the first crystals."

And here is the significant statement, Your Honor:

> "It appears therefore that the qualifying term 'about'" -- we

have a term here "about 355 degrees Centigrade" --

> "is intended to include a variation of up to 5° above or below the figures specified."

And they state:

> "We do not believe that it can be seriously asserted that a melting point range as wide as indicated defines an identifying characteristic."

The Board was referring to the well-known property of chemical compounds of having definite melting points and referring to the fact that the purity of a compound will be indicated to a large degree by its melting point. In other words, it is well known and I believe the witness will help us on that, that if we have a compound that has a certain stated melting point and we make a number of determinations of that compound and we don't get any variation from that, we have a pretty good indication that we have a pure compound. On the other hand, if we have melting points going all over a particular range, it is an indication of impurity and is not very much help in indicating what particular compound we may have or indicating a specific melting point that somebody else may use as a basis for determining whether or not he is infringing the
20 claims.

Now, the second point that the Board noted was a reference to an "approximate molecular formula of $C_{10}C1_{10}0$. In other words, ten atoms of carbon, ten atoms of chlorine, and one atom of oxygen. As the Board pointed out and as counsel for the plaintiff also indicated, chemical compounds have a very definite molecular composition. A compound made today and a compound made ten years from hence at some distant point, will have, if they are the same compound, exactly that combination of atoms. We have combined in this empirical or molecular formula, indicated by $C_{10}C1_{10}0$, some combination of these elements; what this combination may be is a critical matter, though, because frequently such a molecular formula may refer to more than one given compound.

To take a simple example, Your Honor, take C_4H_6 -- I am forgetting my chemistry (placing examples on the blackboard), but suppose we have four carbons in a line, we may have a butane radical, butane compound. These carbons can be arranged in this manner (indicating) with two carbons appearing attached to one of the other carbons, we still have four but we have a different compound with different properties. In other words, you have them lined up sometimes, you can have them with branches, you can have a five-sided figure -- I am straying from the butane compound, going into other forms of compounds, this would
21 have five carbons and this could be five carbons in a five-sided figure or we could have, for example, the branched structure with five carbons also.

Chemistry permits an infinite number of compounds, I would say not even explorable by the human imagination. Of course, a mathematician in any one of these simple cases as with butane or with a cyclopenta- compound like cyclopentadiene, which would be the five-sided figure, could give you the number of possibilities involved. As the plaintiff's counsel has stated, the formula has not been suggested for the particular compound here involved.

But my comments have been a digression, in effect, because the Board pointed out that as to this $C_{10}C1_{10}0$, this empirical or molecular formula cannot be considered as significant because it wasn't actually originally disclosed. As the Board pointed out in this case, if this formula $C_{10}C1_{10}0$ is of any significance, it is new matter in this case because the application as filed described this compound as having a molecular formula of $C_{10}C1_{10}$ two oxygens and a hydrogen. In other words, it had an additional hydrogen and an additional oxygen and the formula was changed in the course of the prosecution of the case to its present form. No doubt the Examiner considered this thing of no significance and therefore made no ruling on this point, but as the Board said, if this is significant, it is new matter in the application.

22 Two other points of distinction or possible points of distinction were urged, one, a behavior as a weak organic acid. I do not think that the Board understood this to be an indication that this was necessarily a weak organic acid, but the claims of the application would admit of such an interpretation. The Board's view was an expression like "weak organic acid" or "behaves as a weak organic acid" is too inclusive and indefinite an expression. In other words, it includes too many diverse compounds and is no indication of particular chemical structure or it is very weak identification.

To illustrate this matter of behavior, when you speak of a class of weak organic acids, you are including a large number of compounds and, supposing that that is a definite expression, we don't know which of these items of behavior are selected as identifying features.

Reference has been made to the ionic strength and I will not try to explain ionization because I am not too sure that I know what it is, myself. But there are other items of characteristics, you might say, of a weak organic acid and the claim gives one no identification of the particular characteristics involved.

A third item refers to "very soluble in polar organic solvents" and "substantially insoluble in non-polar organic solvents." I would not call it a guess to state that there are thousands of compounds that could be similarly described, assuming that there was some agreement

23 on what these terms meant, polar organic solvents and non-polar

organic solvents, and also some agreement that these criteria were of any importance.

Now, those are the views of the Board as to claim 6.

Claim 13 imports another matter. Apparently the applicants here were uncertain that they could rely on this collection of unrelated characteristics as a means of identifying this compound, so they combined this means of description with a reference, an indefinite reference to the manner in which this compound might be made. But this reference does not follow the usual pattern in this matter, it does not state that this compound was prepared by a particular process. In other words, the applicants wanted to have their cake and eat it, too.

Now, it has been held in a number of courts that when a man defines his product, chemical product or sometimes a mechanical product, in terms of a method of making that product, he is limited in his protection to just that product, the product of that particular process. This claim goes beyond that concept. It is suggested that this compound, referring to claim 13, is preparable in a certain manner; in other words, it can be prepared in that manner, possibly in many other manners, but the applicant wishes to get the protection on this compound without doing the work of identifying the compound, but requiring his would-be
24 infringer or possible infringer to go to the trouble of identifying the compound.

For example, if some manufacturer was uncertain as to the infringement of a certain one of his products, he could not determine whether he was infringing this patent claim, if such patent claim became a patent claim, unless he carried out this reaction; in other words, he prepared it by the method specified in the claim and he identified that product and then he went further and he carried it out by his own process, perhaps being uncertain as to the identity, and then compared those two products. In other words, he would have to make the determination that the applicant here was unwilling to make.

By the rule of law followed in this Circuit, an applicant in defining his compound, cannot describe it as being the product of a particular process unless there is no other means of doing it. So that in claim 13, we have two factors, we have an identification or an attempted identification of the compound by the characteristics of approximate melting point and approximate molecular formula, and also an attempt to define it by the process of making it, and I believe it would be clear that such a claim would be improper to be considered in this Circuit.

MR. LAURENCE: May I have a short comment on what counsel said?

THE COURT: If you wish to, but as I understand, you are going
25 to file briefs anyway.

MR. LAURENCE: Yes, I just wanted to correct something here. I believe counsel meant -- I believe you meant up here, not an "H" but a "10", is that right?

MR. BEHRENS: I was writing in a hurry.

MR. LAURENCE: C_4H_{10} for a butane. I just wanted to point out while we are both here, that counsel selected butane and I looked in the Handbook because he was complaining bitterly about the fact that 355 wasn't significant on account of you might be five degrees one way or the other.

Now, the two butanes which he selected here, this is the normal form of the butane where the carbon atoms are in straight chain and this is the isoform where two carbon atoms are branched, and the melting points of those compounds, as given by the Handbook, are 21.3 degrees apart. The actual melting points are -138.3 and -159.6, which is considerably more than what we have.

The other example he went to, cyclopentane and normal pentanes, I find here that normal pentane melts at -129.7, the iso or branched chain form, -159.9, which is thirty degrees apart; neopentane, -16.6, which would be a variation of some 113 degrees from the normal; and then he went to the ring, and that is cyclopentane which melts at -93, it is about 36 degrees off the normal form.

So, in every one that he picked out, the variation is far greater,
26 many, many degrees greater than the variation which he complains of in our case as being insufficient to be a fingerprint.

Mr. Sconce, please.

Thereupon --

JAMES S. SCONCE

called as a witness by the plaintiffs, being first duly sworn, was examined and testified as follows:

DIRECT EXAMINATION

BY MR. LAURENCE:

Q. Will you please state your name, age and place of residence? A. James S. Sconce, 51 years old, Lewiston, New York.

Q. And your occupation? A. I work for The Hooker Electrochemical Company as Technical Assistant to the management.

Q. Will you very briefly tell us your qualifications as a technical man, starting with your degrees? A. I graduated as a Bachelor of Science from the University of Illinois, entered the field of chemistry in 1928, have been actively engaged in that field in both organic, inorganic, and electro-chemistry since that time. I was Manager of Chemical Research for The Hooker Company for a number of years.

Q. Would you say that you are a chemist both from the stand-
27 point of having worked in the laboratory and also a theoretical chemist? A. Both.

Q. Now, I believe you have heard the opening statement of counsel for the Commissioner of Patents and counsel says that the expression here, which we will call the fingerprint of this compound, as having a melting point of about 355 degrees Centigrade, is meaningless, and relies upon the opinion of the Board of Appeals of the Patent Office which also said:

> "We do not believe that it can be seriously asserted that a melting point range as wide as indicated defines an identifying characteristic."

Now, what statement can you make with reference to that finding of fact by the Board of Appeals, if any? A. This melting point range, so to speak, as taken from Table I, covers a series of compounds; it doesn't cover one compound, it covers a compound and its hydrates, as is clearly stated in the specifications. As you heat a compound that contains water, up near its decomposition temperature, and the melting point of lead is a pretty high temperature, you will find that that compound begins to give off water and go back to the true compound. As a result, you get a mixed melting point of the hydrate and the individual, itself. As a result, you get the first softening point, the first time that you see those crystals begin to crumble and change their physical form, which is shown as the first softening point in that table.

28 Q. May I interrupt you, perhaps it would be helpful to the Court to tell us very briefly how you determine a melting point. A. All right. The melting point of a compound up in that range is determined by taking a small capillary tube, about one millimeter outside diameter and smaller inside, seal off one end, pack it to a depth of about one inch with crystals, then you seal the other end of it because, as I said, the compound will decompose near its melting point. Then you put that into a bath and slowly heat that bath up; you can heat it rapidly until you come near the point where it begins to soften, but from that point on you do it very slowly. The first thing you determine is a first softening point, the point at which the crystans begin to crumble. The second point you determine is the first meniscus, the first time you determine --

THE COURT: The first what?

THE WITNESS: Meniscus, the first time you see any liquid going across the full width of the inside of the tube. Now, then, the mass will gradually go from a solid to a liquid, and then the last point to determine is the last crystal point; in other words, the last time you see a crystal.

All of those points mean something to the analytical chemist who is trying to determine melting points, but he accepts as a melting
29 point range the zone between, as a rule, the first crystal point and the last crystal point, and if there is a range in which 90 or 95% of that melts in that period, in other words, goes from a solid to a liquid, that is generally accepted as a true melting point figure.

Now, for example, we sold a great deal of hexachloroethane from 1942 to 1945 for war purposes. That compound melts and sublimes at 186 degrees Centigrade; as a result, it had to be a sealed melting point determination, yet we set up government specifications with that and used that sealed tube melting point procedure as a standard procedure of determining melting points.

BY MR. LAURENCE:

Q. What would be your view as to the accuracy of a melting point determination with reference to where the melting point is on a scale, say running from room temperature, which is about 23 or 24 degrees Centigrade, up to temperatures such as 335 degrees Centigrade? A. In our laboratory work at Niagara Falls, we determine temperatures in the neighborhood of 20 to 50 degrees Centigrade accurately to a hundredth of one degree. When we get up to 335 or 355 degrees Centigrade -- I believe that should be 355 (indicating figure on the blackboard).

Q. Perhaps it is (making correction on the blackboard). A. -- temperature is a much wider figure, two degrees, three degrees or four
30 degrees, is quite significant to us.

Q. Are you familiar with a book called "Organic Chlorine Compounds" by Huntress, which was published about 1948 by John Wiley and Sons? A. Yes, about half of our chemical business is on chlorinated compounds, most of which are listed in that book.

Q. About how many compounds are listed in this book? A. Oh, about 1300, somewhere in that neighborhood.

Q. Now, have you recently examined the book, which I believe is pretty well indexed, and ascertained the compounds which contain either carbon, chlorine and oxygen, or carbon, chlorine, hydrogen and oxygen? A. Yes, I have gone through that very carefully.

Q. Will you tell me the melting point of the compound which is closest to 355 degrees Centigrade and which has the empirical formula of the claim or the empirical formula which counsel for the Solicitor's Office has placed on the board? A. There is no compound in that book having a molecular formula $C_{10}Cl_{10}O$, $C_{10}Cl_{10}HO_2$ or $C_{10}Cl_{10}H_2O_2$.

Q. All right. Now, since you can't find those compounds in the book, will you tell me what came the closest to it and what its melting point was? A. The nearest compound containing those elements is pentachloropropionic acid, $C_3HO_2C1_5$. The melting point of that compound is listed by Huntress as 200 to 215 degrees Centigrade,
31 a range of 15 degrees.

Q. Could you find any compound in which in any permutation of C, C1, O or C, C1, O and H, there was a melting point within 50 degrees Centigrade of 355 degrees Centigrade? Would you like the question read? A. I am thinking back one minute to that previous statement. There is one limitation in that, Mr. Laurence, that contain from 68 to 73% chlorine.

Q. Yes. A. Of that previous compound.

Q. Yes, that is within this ratio. A. That is within that molecular range.

Q. Right.

THE WITNESS: Would you read the question back?

(The pending question was read.)

THE WITNESS: Not if they contain from 68 to 73% chlorine.

BY MR. LAURENCE:

Q. And the compound which is specified on the board as $C_{10}C1_{10}0$ does contain within that range of chlorine? A. Yes, that calculates between 71 and 72% chlorine.

Q. Does the formula $C_{10}C1_{10}0$ give to a chemist the molecular weight of the compound? A. Yes, it does, very definitely.

Q. By a simple computation? A. Yes.

32 Q. Now, will you tell me what the highest molecular weight given in Huntress for compounds of this type is? A. 418.6.

Q. And what is the molecular weight of the compound here? A. 493.

Q. Have you examined the standard handbooks such as Lange and the Chemical Rubber Handbook recently in connection with this matter? A. Yes, I have.

Q. About how many organic compounds does each handbook list? A. Well over 7,000 in each handbook.

Q. Is there any compound listed therein which melts at 355 degrees Centigrade, organic compound? A. None exactly at 355 degrees Centigrade.

Q. All right, let's take a range of 5 degrees either way, 350 to 360 degrees, then what do you find? A. In the Handbook of Chemistry and Physics, you will find about 11 compounds. Of those 11 compounds, only one of them contains a halogen and that halogen is not chlorine.

Q. What is it? A. That compound is hexaiodobenzene.

Q. Will you state the empirical formula? A. C_6I_6.

33 Q. What about Lange's book? A. Lange's gives a little different classification. They cover a range of 341 degrees to 360 degrees, a range of 19 degrees Centigrade. In that range, they list 9 compounds only. Again, the only halogen containing compound or chlorine containing is the hexaiodobenzene and there is no chlorine containing compound in that group of 9 chemicals.

Q. Do each of these handbooks have charts which give the melting point ranges of organic compounds? A. Yes, they do.

Q. It was not necessary for you to go through the 7,000 compounds; it is found readily in here and is used by chemists? A. I used their classification, it is standard procedure for chemists. It might be pointed out, as you go into the middle ranges of temperature of 50 and 60 and 70 degrees Centigrade, that for a 5 to 8 degree Centigrade range, you will find as many as 200 compounds in those classes. It is clear that in compounds that melt at 300 degrees Centigrade and higher, there are very, very few listed and known to the chemists.

Q. Now, will you tell us a little bit about the significance of the expression that the compound behaves as a weak organic acid? A. When you take the compound similar to this one, with one oxygen in it, we have to make some assumptions as to its structure. I think it is
34 fairly well accepted by the three groups who have worked very carefully and thoroughly in this field, the Purdue Research Foundation

people, our laboratories, Hooker Electrochemical, and the Allied Chemical people, that that oxygen is there as a ketone, which is a C double bond oxygen group. Ketones have a very common characteristic in forming hydrates with water, which means that they react in a weak manner with water. We found that when this ketone reacts with water, it does put hydrogen ions into solution and that those hydrogen ions can be titrated by normal procedure with caustic soda or caustic potash, just as any weak organic acid.

We also find another characteristic of the behavior of weak organic acids and that is that they will react with amines and with other basic compounds. The hydrogen ion will attack and form salts or esters.

Q. I have here McElvain's book on "The Characterization of Organic Compounds," which was published by the McMillan Company in 1945. What comment can you make as to how this book is regarded by chemists in the field of characterizing organic compounds? A. Professor McElvain is one of the leading scientists in the United States. I believe he is head of the Organic Chemistry Department at the University of Wisconsin.

Q. Now, in this book at page 2, there is given an outline of procedure which has to do with a characterization procedure. Will you refer to page 2 and tell us what it says with reference to melting point as an index or characteristic aiding or assisting in the determination of a specific compound? A. The melting point of a solid and a combination of the boiling point and the refractive index of a liquid are the most used physical properties for the characterization of organic compounds.

Q. The compound we have here is a solid, right? A. This is a solid, not a liquid.

Q. Therefore, based on that, you find that McElvain says that the melting point is the most useful characteristic of all, correct? A. That is correct and our handbooks also bring out that point very clearly by listing melting points as one of the most important and practical factors.

Q. I believe you were present and heard me read from the handbook the identifying headnotes giving each type of property listed in the handbook for determining or identifying chemical compounds, is that correct? A. Yes.

Q. Was my statement to the Court correct? A. Yes, that is correct.

Q. Now, will you tell us a little about the significance of the
solubility characteristic? Does that have any meaning? A. Yes, it
36 means a great deal. Compounds that are polar are usually
soluble in polar solvents and insoluble in non-polar solvents, and vice-
versa, compounds that are non-polar are usually soluble in non-polar
solvents and not as soluble in the polar solvents. Now, in this case,
you have certainly a polar compound. It does dissolve quite freely in
polar solvents; it is quite insoluble in the non-polar solvents, such as
carbon tetrachloride and hexaiodobenzene.

Q. Now, let's go to the last identifying characteristic, that is the molecular formula. Is this molecular formula of significance, where the word "approximate" occurs in the definition? A. Yes, molecular formula is again one of the early physical properties that is determined by the average chemist in attempting to identify an organic compound. Carbon, hydrogen and oxygen are usually determined by a combustion analysis, which is a common standard procedure known to all chemists. Most of us can't run them any more, but we do have our analytical people who are skilled in the art and capable of doing that job for us. Chlorine is usually determined in a compound of this sort, by ignition with sodium peroxide or lime, or some such procedure. The accuracy of the methods are complicated when you get into compounds of this sort and they are complicated also by the purity of the original compound.

Now, as I pointed out on this compound to begin with, it forms
37 hydrates, it reacts --

Q. Will you explain for the Court what hydrates are, please? A. A hydrate is addition of water to a compound. I think you are probably familiar with calcium chloride; if you put that out in the air of this courtroom, it would eventually pick up enough moisture from the air to turn itself into a liquid, that liquid is a hydrated form of calcium chloride.

In the same manner, this compound can pick up moisture from the air to form a hydrate and that was one of the reasons that the original formulas, as supplied, contained hydrogen, because water is nothing more than H_2O, which are two hydrogens and one oxygen.

The significance of this 493 molecular weight is quite important. Now, we would expect to determine molecular weight accurately in that range to maybe five or six, or if the true molecular weight is 493, we would be satisfied with a molecular weight determination of 490 or 496, I don't think it would be much more reliable than that.

Q. Suppose you were in a range around a weight of 40 or 50, how accurate could your determination be? A. That would be much better, you would be probably within one unit.

Q. By the way, what is the atomic weight of hydrogen? A. One.

38 Q. And I believe you said the molecular weight of the whole compound is -- A. 493.

Q. So you are hunting for 1 in 493 which would be about, what, two-tenths of one percent or less? A. That is right.

Q. When you are not sure of your analysis on whether there is a hydrogen atom there? A. The hydrogen content is nineteen-hundredths of one percent.

Q. There is an additional oxygen atom, which is 16, applied against 493 and what is that, percentagewise? A. About three-tenths.

Q. Of one percent? A. Yes. I am sorry, three percent.

Q. Three percent, not three-tenths, all right. I think I will just ask one final question. From your experience and knowledge, does or does not this combination of characteristics, as set forth in claim 6, accurately define a single chemical compound and only one? A. That is correct. It does.

MR. LAURENCE: You may cross-examine.

THE COURT: We will take a short recess.

(A short recess was taken.)

MR. LAURENCE: The Solicitor and I have agreed, instead of
39 putting specific items in evidence, we will introduce the books and with the permission of the Court, we may each use photocopies of such pages as we wish to in connection with the briefs. Do you want to list them?

THE COURT: Do you want each book marked separately? I suppose they should be.

MR. LAURENCE: Yes, I think we should.

Why don't you just take them in order and list them, you can do that at your leisure and I will go on.

THE COURT: Let's get them marked.

DEPUTY CLERK: Handbook of Chemistry and Physics will be marked as Plaintiff's Exhibit No. 3.

THE COURT: Do you have any objection to the book, itself, being marked?

MR. LAURENCE: No, none at all.

(Handbook of Chemistry and Physics, 39th Ed., by Chemical Rubber Publishing Company, was marked Plaintiff's Exhibit No. 3 for identification and received in evidence.)

DEPUTY CLERK: The Characterization of Organic Compounds by McElvain, I guess published by McElvain and written by McElvain, will be marked as Plaintiff's Exhibit No. 4.

(The Characterization of Organic Compounds by McElvain, was marked Plaintiff's Exhibit No. 4 for identification and received in evidence.)

40 DEPUTY CLERK: Handbook of Chemistry by Lange, the 8th Edition, will be marked as Plaintiff's Exhibit No. 5.

(Handbook of Chemistry, 8th Ed., by Lange, was marked Plaintiff's Exhibit No. 5 for identification and received in evidence.)

DEPUTY CLERK: Organic Chlorine Compounds by Huntress, will be marked as Plaintiff's Exhibit No. 6.

(Organic Chlorine Compounds by Huntress, was marked Plaintiff's Exhibit No. 6 for identification and received in evidence.)

DEPUTY CLERK: The Encyclopedia of Chemistry by Clark and Hawley, will be marked as Plaintiff's Exhibit No. 7.

(The Encyclopedia of Chemistry by Clark and Hawley, was marked Plaintiff's Exhibit No. 7 for identification and received in evidence.)

THE COURT: Very well, they are received. Did you have anything further?

MR. LAURENCE: No further questions, Your Honor.

CROSS-EXAMINATION

BY MR. BEHRENS:

Q. Perhaps it is my perverse sense of humor, but how many times has your company written out an order for organic chemicals in quantity and used a designation such as the items listed on the board here, crystalline solid, melting point of about 355 degrees Centigrade, behaves as a weak organic acid, very soluble in polar organic solvents and substantially insoluble in non-polar organic solvents, and an approxi-
41 mate molecular formula of something or other; could you answer that? A. We wouldn't purchase under that class of specifications. We would write out those specifications into a chemical specification and give it a number. We sell many compounds under code numbers that we cannot identify properly by anything other than a chemical description of that nature. I think we actually have sold some pounds of the parent compound under the trade name JN-63, using the initials J and N for Jack and Newcomer, the inventor of the compound. That was the best we could define it, plus the information as you saw it in the JN-63 patent.

Q. In other words, you were putting your corporate reputation for honesty behind some particular designation by a number? A. That is correct.

Q. Now, I hand you U.S. Patent 2,616,825 to Gilbert, et al. and I ask you to turn to column 4 of that patent and look at the type formula or the structural formula in the middle of that column. Would you say whether that is the same compound that we have involved in this case? A. You mean reading from the structural formula?

Q. Yes, that is right, or looking anywhere in the specification of that patent. A. I don't think it is my belief or the belief of any of
42 our people at The Hooker Company or the belief of the Purdue people, who have published a paper on the subject, that this compound exists.

Q. It, however, would respond to the molecular formula that is given here, would it not? A. It would respond to the molecular formula.

Q. It was not found in the Handbook and the book by Huntress, to which you referred in your testimony? A. That is correct.

Q. It was not? A. It was not.

Q. Could it be that there are numerous other compounds or perhaps one or two other compounds described in the literature -- let me change that, undescribed in the literature but perhaps described in an application in the Patent Office, which would have the same molecular formula that you have in this compound here? A. Yes, very definitely.

Q. Isn't it true that we could also assume that perhaps there are compounds being described in patent applications in the Office and not yet accessible to the public, having melting points very close to this range, very close to this 355 degrees that we identify as about 355 degrees here? A. You mean that there may be other compounds that have a melting point near 355 degrees Centigrade, other than the compound
43 here?

Q. Yes, that is right. A. Yes, there can be other compounds.

Q. In other words, those tables are significant only as to the compounds which are well known already to the art; I will amplify, the tables in the Handbooks and in the books which you referred to in your direct testimony? A. No. Huntress, for example, made a rather thorough search. Beilstein's Handbook lists up through 1929, I think, almost every chemical compound known at that time of the organic chemical compounds. Dr. Huntress, in developing the Group 3, a compound containing chlorine, carbon and oxygen, reviewed all of Beilstein, all of the Beilstein supplements, and something like 22, 000 other literature references containing chlorine compounds. That list is probably the most complete in the world. It contains not only common compounds but it contains every uncommon chrlorine compound that Dr. Huntress, in two years of literature search could find.

Q. Is it not true that these compounds were known to the art before, though? In other words, when we speak of Beilstein, is it not merely a compilation of known compounds? A. It is a compilation of all known compounds in as far as they were able to search the literature, which is very, very complete. Beilstein and Huntress, between them, have done a remarkable and fabulous job.

44 Q. But this is no guide in talking about newest developments, is it? A. The research work on this process was done back in 1946 and 1947. Dr. Huntress published that book, I believe, in 1948 and the book was written and turned over to the publishers in 1946. Therefore, it should be very nearly up to date for chlorine compounds known of that period.

Q. I will hand you a photocopy of the title page of a standard organic work, A. F. Holleman's "Organic Chemistry" with a translation by J. P. Wibaut. Are you familiar with this work? A. I am not familiar with this text, no.

Q. You will notice on the title page that it is translated from the sixteenth Dutch edition by Samuel Coffey, Section Head, Research Department, I. C. I. Dyestuffs Division. Would you not assume that a textbook that went to sixteen editions would be somewhat well liked in the scientific field? A. I imagine it is a very good book.

Q. I will hand you, if you will look, the excerpts that I hand you include pages 29 and 30, and I have marked on page 30, underlined in red, a sentence from this textbook. Would you please, for the benefit of the Court, read that short sentence and state whether you believe it

to be accurate? A. "In the practice of organic-chemical research it is
in many cases sufficient to determine the melting point with an accuracy
45 of 0.5 to 1.0° C. 10-20 Milligrams ..." Now, that is correct, as
I have previously stated, in the range of normal melting points. It is
not correct in compounds in the melting point range of 360 to 340 Centigrade, what we are talking about in compounds that decompose near their melting point.

Q. Please speak to the Court. A. Now, the example I gave earlier on hexachloroethane where the compound sublimes at its melting point and is a high melting point where you have to go to this sealed tube capillary procedure, is a good example of why this doesn't hold true when you get out to the outer edges. This would be fine for a chemical text for normal reasons. It isn't true when you get up to the melting point of lead.

MR. BEHRENS: I ask that this excerpt from Holleman's "Organic Chemistry" be admitted in evidence as Defendant's Exhibit 1.

THE COURT: Any objection?

MR. LAURENCE: No, Your Honor.

THE COURT: It will be received.

(Pages 29 and 30 from Holleman's "Organic Chemistry" were marked Defendant's Exhibit No. 1 for identification and received in evidence.)

BY MR. BEHRENS:

Q. I hand you photocopies of a title page and pages 21, 22 and
23, taken from the textbook, "Practical Organic Chemistry" by Arthur
46 I. Vogel, Doctor of Science of London. Could you tell me
whether you are familiar with this textbook? A. I have not used it.

Q. Would you please look at page 21 and read the underlined sentence at the bottom of the page? A. "For pure substances, the change from the solid to the liquid state is quite sharp (within five-tenths of a degree), hence the temperature is valuable for purposes of identification."

MR. BEHRENS: I ask, Your Honor, that this excerpt, title page and pages 21, 22 and 23 from the textbook of "Practical Organic Chemistry" by Arthur I. Vogel, be admitted in evidence.

THE COURT: It will be received as Defendant's 2.

(Pages 21, 22 and 23 from Vogel's "Practical Organic Chemistry" were marked Defendant's Exhibit No. 2 for identification and received in evidence.)

BY MR. BEHRENS:

Q. I don't want to overdo this, but I will include one more excerpt. I will hand you the title page and pages 23 and 24 of the book, "Laboratory Technique in Organic Chemistry" by Avery Adrian Morton, Associate Professor of Organic Chemistry at the Massachusetts Institute of Technology. Are you familiar with the work or with the man? A. Yes, I am familiar with this.

Q. Would you please turn to page 24 and read the underlined sentence? A. "Tseng reports discrepancies that amounted to more
47 than 2° with different types of apparatus and differences of as much as 1° with different observers using the same apparatus. These figures represent probably the maximum amount of divergence." He is there, as I remember, speaking about the differences in melting points by using different types of apparatus in a reasonable range of temperature.

MR. BEHRENS: I request, Your Honor, that this be entered in evidence as Defendant's Exhibit 3, I believe.

THE COURT: It will be received.

(Pages 23 and 24 from Morton's "Laboratory Technique in Organic Chemistry" were marked Defendant's Exhibit No. 3 for identification and received in evidence.)

BY MR. BEHRENS:

Q. With reference to your statement on the significance of solubility in polar and non-polar solvents, I hand you the title page and pages 575 and 576 of the book, "Natural and Synthetic High Polymers" by Kurt H. Meyer, Ph.D. Are you familiar with Dr. Meyer and his work? A. Yes, I am.

Q. Would you not say that he is one of our -- I should say was, I believe he died recently -- greatest scientists? A. He was an expert in the polymer field.

THE COURT: In the what field?

THE WITNESS: Polymer chemistry field.

48 BY MR. BEHRENS:

Q. I point to page 576 of that excerpt. Would you read, please, from the underlined subject matter? A. This is an excerpt from the work on polymers in the influence of individual groups on solubility. Again, this is limited to speaking of polymers; I don't think we are speaking of polymers in this case. However, I quote:

> "On the other hand, the search for a relationship between solvent powers and dipole moment of the solvent appears to be hopeless, since both substances with a polar, hydrophobic groups and those with hydrophilic groups may, in spite of profoundly different solvent powers, possess the same dipole moment."

MR. BEHRENS: Your Honor, may I ask that this be introduced in evidence as Defendant's Exhibit 4.

THE COURT: Very well, it will be received.

(Pages 575 and 576 from Meyer's "Natural and Synthetic High Polymers" were marked Defendant's Exhibit No. 4 for identification and received in evidence.)

BY MR. BEHRENS:

Q. Now, this classification of dipolar or polar and non-polar, intrigues me. Apparently Dr. Meyer did not consider it of significance from the solvent viewpoint. A. Not in polymers.

Q. Now, is benzene, our most common organic solvent, polar
49 or non-polar? A. Its dipole moment is about 1.003, which
makes it almost non-polar.

Q. Almost non-polar? A. Yes.

Q. Which would you put it in, this category here? A. Non-polar.

Q. Non-polar. Would you put toluene, which is its nearest relative, you might say, in the chemical field, in the same group? A. There you have gone up the dielectric constant scale and it has more polarity and you are beginning to get into something that might be a borderline polar solvent.

Q. Now, you have given me two of the more common solvents and we have had two borderline cases; is that typical of this method of identification? A. You mean benzene is a borderline case as a non-polar solvent? No, I did not say that. I said benzene is as close to being a non-polar solvent as it possibly could be.

Q. But it is polar? A. All solvents have a degree of polarity, as far as I know.

Q. In other words, if they all have a degree of polarity, then practically all solvents could be said to be polar to some extent?

A. No. There is a very definite understanding, as chemists use that term, and probably any solvent that is under 1.005 in dipole moment would be considered as being non-polar by the average chemist. If you want to be completely and thoroughly technical, about the only polar solvent could be water at very specific temperature pressure and no specific degree and condition.

Q. There is just one more matter and that is this question of "behaves as a weak organic acid." Don't organic acids have many points of behavior, in other words, aren't there many different things that you can say are common to weak organic acids? A. They will do many things that are common; we have recited a number of them in the patent application such as the forming of salts, such as reaction with alkalis, such as reaction with amines. A list was given in the patent specification of the things that they do where they are comparable to --

Q. But do not these weak organic acids differ among themselves in various significant characteristics? A. Certainly.

MR. BEHRENS: That is all, Your Honor.

REDIRECT EXAMINATION

BY MR. LAURENCE:

Q. I believe counsel for the Commissioner showed you a copy of
the Patent 2, 616, 825 and you were substantially his witness on the mat-
51 ter because it was not brought out in the direct examination. I
would ask you this, whether that patent is the so-called use patent of the
compound which was patented to Gilbert, one of the co- --

MR. BEHRENS: Objection, Your Honor. The witness had not qualified as an expert on law or interpretation of patents.

MR. LAURENCE: I am not going to ask him a legal question.

THE COURT: Proceed, finish your question.

MR. LAURENCE: Will you read the question?

(The question was read as follows: "I believe counsel for the Commissioner showed you a copy of the Patent 2, 616, 825 and you were substantially his witness on the matter because it was not brought out in the direct examination. I would ask this, whether that patent is the so-called use patent of the compound which was patented to Gilbert, one of the co-"

BY MR. LAURENCE:

Q. -inventors in Patent 2, 702, 305; that is, the patent the Solicitor showed you calls for the pesticide use of the compound which is covered in the 2, 702, 305 Patent, is that correct? A. Claims 1 and 2 in front of me call for a pesticidal composition.

Q. Now, the Patent Office is peculiarly in possession of some
information which I would not have and I may want to inquire about it.
52 The compound which is covered in the patent shown you by the
Solicitor and in the patent which I have shown you, 2, 702, 305, is the
same compound; is that correct? A. The structural formula as given
is the same.

Q. Now, I will state and I believe the Solicitor will not contradict me, that the compound Patent 2, 702, 305 was reissued as Patent 24, 397 on November 26, 1957.

MR. BEHRENS: I have no knowledge of either patent on that.

(The patent was handed to Mr. Behrens.)

MR. BEHRENS: You have the advantage of me.

MR. LAURENCE: Well, will you agree that -- you brought this up, I didn't -- will you agree that the patent was reissued, that this is a bona fide copy of the United States Patent?

MR. BEHRENS: I am not challenging any statement of yours.

MR. LAURENCE: Perhaps I can ask the witness or you, the fact is that the formula to which counsel made reference has been deleted throughout the compound patent?

MR. BEHRENS: I beg your pardon, I believe we are talking about two different things. This says the original 2,702,305 and as I remember, the patent to which I referred was 2,616,825.

MR. LAURENCE: I agree with that, but I don't know whether
that patent is in the process of being reissued or not, that is why I said
you have the advantage. I can only point out that the formula which occurs
53 in the use patent that you gave the witness, has been deleted by
reissue in the compound patent of the same inventor.

MR. BEHRENS: May I point out that that is a different compound claimed there.

MR. LAURENCE: All right, let's ask the witness about that, because the witness said it was the same. Now, which particular formula did you make reference to in the 2,616,825 patent?

(Mr. Behrens indicated the formula.)

MR. LAURENCE: All right, that is the decachlorotetrahydro-4, 7-methanoindeneone compound about the center of column 4 of the 2,616,825 patent, is that correct?

MR. BEHRENS: So it is identified.

MR. LAURENCE: I will circle that in red in the copy of the patent here.

BY MR. LAURENCE:

Q. Now, Mr. Sconce, will you look at the formula concerning which I have just had a discussion with counsel and tell me whether or not that is the compound by the same formula which was deleted by the same applicant Gilbert from his compound per se patent, when he reissued it as No. 24,397 on November 26 of 1957? A. The empirical

formula of both of these compounds is $C_{10}C1_{10}0$, which is identical with the empirical formula on that blackboard. The structural formula which
54 gives a picture of the thing, and is representative to most chemists as having significance, is the same.

Q. What significance would you attach to the deletion of the structural formula and the name of the compound from the compound patent of Gilbert by his Reissue 24,397, speaking as a chemist? A. In preparing the filing of this case, the Purdue people in our laboratory considered very seriously whether we should attempt to include a structural formula of this type in the patent. There was so much question as to the proper structure of that compound that we felt it unwise to try to define that product by a formula which didn't hold together. That has been substantiated and was published in the article that I mentioned a little bit earlier by Dr. McBee in 1955 in the Journal of the American Chemical Society and I would imagine that a deletion from the patent by a reissue would mean that Dr. Gilbert had also arrived at the same conclusion.

Q. In his opening statement, counsel was talking about a determination of infringement of this claim, how a person who was making a compound would know whether he had the compound of the claim, if it be permitted to issue in a patent. And he said, I think I am quoting him accurately, that the purported infringer would have to make a determination the applicant is unwilling to make, unquote. Now, in fact, would not the only thing necessary in a determination of infringement be to
55 take a mixed melting point with a standard sample of the compound as supplied by Hooker and make a determination of the formula? You might explain for the Court what a mixed melting point is, if you please.

MR. BEHRENS: Your Honor, I feel that I have to object to that question. As I stated before, the witness has not been qualified as an expert on patent law and the interpretation of the matter of infringement would be a matter of law, as any bar committee would suggest.

MR. LAURENCE: My question is, first of all, to have the witness tell the Court what a mixed melting point means and then to tell the

Court whether the simple determination of a mixed melting point of any compound, purported to be an infringement, would determine the question of the similarity of the compounds as a matter of scientific fact.

THE COURT: You may answer.

THE WITNESS: A mixed melting point, when you have two unknown compounds that you think may be the same, you determine the melting point on each of the compounds individually. Then you mix the two compounds and find the melting point of the mixture. If you get the same melting point figure, approximately, on all three, you assume it is about the same compound. If the melting point of the mixture is much lower, then you are fairly certain that you have two different compounds.

56 MR. LAURENCE: That is all, Your Honor.

MR. BEHRENS: May I clear up one point before I proceed with the cross-examination. The Patent Office is not relying on the Patent 2,616,825 as an anticipation of this compound here claimed; if it had so intended, the defendant's counsel would have mentioned it in the opening statement. The patent was merely referred to, to suggest one possible compound that might meet this empirical formula; I attached no other significance to it.

MR. LAURENCE: I think perhaps we ought to put in evidence copies of those patents; do you agree?

MR. BEHRENS: It makes no difference to me.

MR. LAURENCE: If you intend to use them to argue what you just said, I think we should put them in. If you intend to drop the whole matter, I am content to drop it.

MR. BEHRENS: Your Honor, I offer in evidence the Gilbert Patent 2,616,825.

THE COURT: It will be received as the next defendant's exhibit.

DEPUTY CLERK: Defendant's Exhibit No. 5.

(Patent No. 2,616,825 was marked Defendant's Exhibit No. 5 for identification and received in evidence.)

MR. LAURENCE: And I will offer as Plaintiff's Exhibit 8, the Gilbert Patent 2,702,305, and the Reissue of Gilbert Patent Exhibit 8, as Exhibit 9, the Reissue being 24, 397 on November 26, 1957.

57 THE COURT: They will be received.

(Patent No. 2, 702, 305 was marked Plaintiff's Exhibit No. 8 for identification and received in evidence.)

(Patent No. Re. 24, 397 was marked Plaintiff's Exhibit No. 9 for identification and received in evidence.)

RECROSS EXAMINATION

BY MR. BEHRENS:

Q. Now, if you were given the job of preparing a chemical compound and you wanted to be sure that it was not the same compound as this compound here claimed, I point to the board with the extracts from claim 6, could you make that determination by merely looking at your own compound? In other words, discovering its melting point and its solubility and polar and non-polar solvents and its behavior as a weak organic acid and its ratio of ingredients, in other words, the percentage of chlorine, percentage of oxygen and percentage of carbon? A. We were given that problem to do. We were given the empirical formula $C_{10}C1_{10}0$ and asked how do you make that compound in other ways than we have produced it? We did that job. We made this compound four different ways in our laboratory; we made the same compound in every case. We made it once, the chlorosulfonic acid reaction on hexachlorocyclopentadiene, and then the hydrolosis.

58 Q. I believe you misunderstand my question. I don't believe it is contradicted that you made this more than one way. Suppose that you were a manufacturer of chemical compounds and you had a new compound that you wanted to put on the market and you didn't know whether or not it infringed this patent claim, we will call it a patent claim. Could you do that, determine whether that compound infringed that claim described by that claim without preparing this compound, I point to the compound on the board and the compound in the application? A. Yes, we could, that is our business. It is that kind of business we are in.

MR. BEHRENS: Thank you.

THE COURT: Anything further?

MR. LAURENCE: That is all. Would you like the briefs at the same time or one before the other?

THE COURT: How long will it take you, do you think?

MR. LAURENCE: How long will the transcript take?

THE REPORTER: Ten days.

MR. LAURENCE: This is January 24, that means about February 3. It depends on the Judge out in Detroit, I have a case in trial out there.

THE COURT: You indicate how long you think.

MR. LAURENCE: I would hesitate to have a brief in before the third week in February. How about around the 24th?

59 MR. BEHRENS: Your Honor please, I believe I have a brief due in this court.

THE COURT: I think you have got a thirty-day limitation now on a brief that is due on that case heard the first of this week, I believe.

MR. BEHRENS: I believe it was last Wednesday, Your Honor. How about February 28th?

THE COURT: Can you exchange and file briefs on February 28th and reply briefs ten days thereafter?

MR. LAURENCE: All right. Thank you.

(Whereupon, the foregoing proceedings were concluded.)

- - -

Plaintiff's
Exhibit 1

APPLICATION

Applicants: Earl T. McBee and
Jack S. Newcomer
For : COMPOSITION OF MATTER

Honorable Commissioner of Patents
Washington 25, D. C.

Sir:

Herewith the above-identified application together with the Filing Fee of Thirty Dollars ($30.00).

Respectfully,
/s/ Dean Lawrence
Attorney

Saint Johns, Michigan
1951 July 21
GWH:sjm:DL 5 CO

COMPOSITION OF MATTER

The present invention relates to a novel composition of matter, more particularly to a chemical compound believed to have the approximate molecular formula $C_{10}HCl_{10}O_2$. While the exact elemental composition and structure of the new compound are not precisely known at the present time, sufficient of its physical and chemical properties have been determined to allow its positive identification and production by any one of several procedures.

It is an object of the present invention to provide a novel and useful composition of matter, viz., a chemical compound, and derivatives thereof. Other objects will be apparent to those skilled in the art to which the invention pertains.

The compound of the present invention has been found to have utility in various ways. For example, it may be employed in its anhydrous form as a desiccant, due to its hygroscopic nature and tendency to form hydrates even under most arid conditions. In addition, the new compound has been found to be a valuable ingredient of insecticidal compositions.

The compound of the present invention, when in the anhydrous state, is a hygroscopic white crystalline solid containing about seventy percent chlorine, having a corrected melting point in a sealed tube of about 355 degrees centigrade, having properties of a weak organic acid, and having the additional chemical and physical properties disclosed hereinafter in the Examples and under "Properties of $C_{10}HCl_{10}O_3$". Due to the broad variety of properties possessed by the compound and its derivatives, it is adaptable to a wide range of utility, of which the capacities mentioned previously are representative.

The compound of the present invention has been described but not claimed in U. S. Patent 2 516 404, issued 1950 July 25 to the present applicants. This patent is concerned with an entirely different invention on another composition of matter, having the approximate molecular formula $C_{10}H_2Cl_{12}O_3S$, which is, however, useful as a starting material in one process for the preparation of the compound of the present invention. In the said patent, the compound $C_{10}H_2Cl_{12}O_3S$ is shown to be preparable by the reaction of hexachlorocyclopentadiene and choloro-sulfonic acid, and the claims of the patent are drawn both to the composition obtained and to the method for obtaining it. Starting with the compound of the said patent, the compound of the present invention may be obtained in a single step or by a simple two-step sequence, as will be more fully disclosed hereinafter.

As stated above, the compound of the present invention may be produced by any one of a considerable number of methods. One very satisfactory method, although not necessarily the preferred method, involves reacting $C_{10}H_2Cl_{12}O_3S$, which can be produced from hexachlorocyclopentadiene and chlorosulfonic acid, with a basic substance, for example inorganic bases such as metallic hydroxides, especially alkali metal hydroxides, e.g., sodium or potassium hydroxide; metallic oxides; ammonia or ammonium hydroxide; organic nitrogen bases such as alkylamines,, e.g., methylamine, diethylamine or triethylamine, pyridine, aniline, phenylhydrazine, or any other primary, secondary, or tertiary amine, whether aliphatic, alicyclic, heterocyclic, or aromatic, or the like. When employing a basic reagent in the preparation of the compound of the present invention, the compound may be recovered as a salt or it may be treated with a mineral and separated as the free acidic compound.

The compound may also be prepared by reaction $C_{10}H_2Cl_{12}O_3S$ with an alcohol, such as methyl alcohol, ethyl alcohol, propyl alcohol, benzyl alcohol, or the like, or with an alcoholic caustic solution. In addition, the compound may also be prepared by reacting $C_{10}H_2Cl_{12}O_3S$ with inorganic iodides such as potassium or sodium iodide.

A more surprising method for preparing the compound of the present invention comprises treating $C_{10}H_2Cl_{12}O_3S$ with activated carbon.

Still another and a preferred method for preparing the compound of the invention comprises treating hexachlorocyclopentadiene with sulfur trioxide and then treating the product with a chemical reagent containing active hydrogen atoms, such as an alcohol, e.g., methyl alcohol or ethyl alcohol; or an acid, such as acetic acid; or even by treatment with water. The reaction conditions such as temperature, ratio of reactants, reaction medium, or reaction time maintained during this preparation are not critical, but it is desirable to effect the reaction of hexachlorocyclopentadiene and sulfur trioxide at a temperature between twenty and sixty degrees centigrade, although temperatures as low as minus twenty and as high as one hundred and twenty degrees centigrade may be successfully employed. The chemical intermediate, which results from reaction of hexachlorocyclopentadiene with sulfur trioxide, may be treated for production of the compound of this invention either in situ or after separation. On the other hand, this intermediate may be subjected to a treatment with hydrogen chloride or even chlorosulphonic acid to produce the compound $C_{10}H_2SCl_{12}O_3$, which is the subject of the above-mentioned United States Patent.

PROPERTIES OF $C_{10}HCl_{10}O_2$

This novel composition of the present invention exists in the anhydrous form as well as in the form of various hydrates. The compositions, both hydrous and anhydrous, are odorless white solids which melt in a sealed tube at about 355 degrees centigrade (corrected). The anhydrous solid is in the form of transparent crystals, whereas the hydrated solids have an opaque appearance. The anhydrous form has a tendency to acquire a marked electrostatic charge. The compound $C_{10}HCl_{10}O_2$ behaves as does a weak organic acid in that it gives an indefinite end-point with phenolphthalein, but a definite end-point with thymolphthalein. Determinations of the neutral equivalent of the material are dependent upon the state of hydration, which in turn is dependent upon the means utilized for purification. The crude solid, as obtained by precipitation from aqueous solution, retains a considerable quantity of free water on "drying" by exposure to atmospheric conditions for several days. The resulting material is very soluble in polar organic solvents such as acetone, methyl alcohol, ethyl alcohol, acetic acid, and is insoluble in non-polar organic solvents such as carbon tetrachloride and benzene. However, if the solid is heated with carbon tetrachloride or benzene to effect an azeotropic distillation of water, a considerable quantity of water of hydration is removed. The hydrated forms have also been dehydrated by recrystallization from acetyl chloride and thionyl chloride as well as by heating to constant weight at 110 degrees centigrade. A dehydrated form, obtained by recrystallization from acetyl chloride and maintained in a desiccator over concentrated sulfuric acid, was found to have a neutral equivalent of 513 and to contain 70.9 percent chlorine. A dehydrated form, obtained by recrystallization from acetyl chloride and kept in a desiccator over phosphorus pentoxide, was found to have a neutral equivalent of 500 and to contain 71.8 percent chlorine.

A dehydrated form obtained by recrystallization from thionyl chloride and kept in a desiccator over phosphorus pentoxide was found to have a neutral equivalent of 478 and to contain 71.5 percent chlorine. Another sample, dehydrated by heating to constant weight at 110 degrees centigrade was found to contain 70.2 percent chlorine. If the anhydrous form is allowed to stand at room conditions, it gains an amount of water equal to approximately 13.1 percent of its original weight. The resulting hydrate has a neutral equivalent of about 588. The hydrate, the anhydrous form, and mixtures of the hydrate with the anhydrous form, all give the same melting point in a sealed tube of approximately 355 degrees centigrade corrected. Some of these data are collected in Table I infra.

Plaintiff's
Exhibit 1

TABLE I[†]

Properties of $C_{10}HCl_{10}O_2$

Treatment	Product Cl. %	N. E.	St. Gain, %	*Melting point, °C. (s. t. corr.)		
				1st soft	1st meniscus	1st crystals
Acetyl Chloride, over H_2SO_4	70.9	513	---	349	351	352
Acetyl Chloride, over P_2O_5	71.8	500	---	---	---	---
Acetyl Chloride, air exposed	---	580	13.1	---	---	---
Thionyl Chloride, over P_2O_5	71.5	478	---	---	---	---
CCl_4, limited air exposure	---	568	---	345	351	355
CCl_4, dried at 110°C.	70.2	526	-7.75	350	355	359
CCl_4, air exposed	---	---	3.7	---	---	---
Acetone, CCl_4, air exposed	64.2	588	---	348	352	355
Methanol, CCl_4, air exposed	---	---	---	351	352	353
Acetic acid, air exposed	---	---	---	331	353	361

† See Examples for additional data.

* None of the materials produces a depression when tested in mixed melting point determinations.

The compound of approximate formula $C_{10}HCl_{10}O_2$ reacts with basic reagents to form salts. The data presented in Table II provide additional bases for assigning the approximate molecular formula $C_{10}HCl_{10}O_2$ to the new composition of matter.

TABLE II

REACTIONS OF $C_{10}HCl_{10}O_2$ WITH BASIC REAGENTS

Basic Reagent	Product % Cl. found	% Cl. theory	m.p., °C. (corr.)
Pyridine	60.3	60.4	143-145 (s.t.)
Diethylamine	61.2	61.1	dec.
Phenylhydrazine	56.3	57.6	160-161

The salts of the compound $C_{10}HCl_{10}O_2$ vary somewhat from each other in solubilities and stabilities. Thus, the salts formed by employing metallic hydroxides, e.g., sodium and potassium hydroxide, are more soluble in water, although only slightly soluble, than is the salt prepared by utilizing phenylhydrazine as the basic reagent. Most of the salts are very soluble in methyl and ethyl alcohols and are relatively insoluble in carbon tetrachloride and benzene. Methyl alcohol, or combinations thereof with water, is a useful solvent for purification of most of the salts by recrystallization. Whereas most of the salts are relatively stable chemicals, some, such as the pyridinium salt, should be crystallized from a pyridine solution in order to prevent decomposition into the reactants.

The presence of ten chlorine atoms in the compound of approximate formula $C_{10}HCl_{10}O_2$ is indicated by the analyses presented in Table I supra, as well as by the fact that two chlorine atoms are removed from the compound $C_{10}H_2Cl_{12}O_3S$ when it is converted to the compound $C_{10}HCl_{10}O_2$. These data are shown in Table III.

TABLE III

CONVERSION OF $C_{10}H_2Cl_{12}O_3S$ to $C_{10}HCl_{10}O_2$

Reagents	Cl-, water soluble, %	2Cl-, theory, %
CH_3OH, KOH	11.53	11.29
C_2H_5OH, KOH	11.34	11.29

Sulfur determinations on the compound $C_{10}HCl_{10}O_2$ have consistently shown 0.0 percent sulfur. The compound of this invention is a relatively stable chemical. It can be boiled with alkaline permanganate and with alcoholic potassium hydroxide without effecting an appreciable reaction.

The following Examples are given to illustrate the practice of the present invention, but are in no way to be construed as limiting.

Example 1 Preparation of $C_{10}HCl_{10}O_2$ from C_5Cl_6:
Step (A) $SO_3 + C_5Cl_6$

Hexachlorocyclopentadiene (2730 grams) was added drop-wise during a period of 3.2 hours to sulfur trioxide (1799 grams) as the temperature of the exothermic reaction increased to 56 degrees centigrade. The temperature was then maintained at 55-61 degrees centigrade for an additional three hours. The gases, which were liberated during the reaction, were collected in a Dry Ice trap, and were found to consist of sulfur dioxide (136 grams) and entrained sulfur trioxide (66 grams). The thick red-colored liquid reaction mixture weighed 4319 grams. An aliquot of 1289.5 grams was distilled at atmospheric pressure until the pot temperature reached 100 degrees centigrade, whereafter the distillation was continued at reduced pressure until the pot temperature reached 112 degrees centigrade, and the pressure had been decreased to three millimeters of mercury absolute. Rectification of the distillate gave 166.5 grams sulfur trioxide and pyrosulfuryl chloride (b.p. 55-58 degrees centigrade at 14 millimeters of mercury absolute). Upon cooling the pot residue to room temperature, the product solidified as a tan solid. Upon cooling the pot residue to room temperature, the product solidified as a tan solid. Upon adding a portion of this solid to water together with a portion of the original crude reaction mixture and then filtering off the solid, virtually no unreacted hexachlorocyclopentadiene could be recovered. The pot residue was recrystallized from a mixture of acetyl chloride and hexane to give a white solid, melting point (corrected, sealed tube) of 349-352-361 degrees centigrade.

Example 2: Step (B) Hydrolysis of C_5Cl_6-SO_3 Reaction Product

A 500-gram aliquot of the crude reaction mixture (totaling 4319 grams, prepared as in Example 1), was added with stirring to three liters of cold water. The temperature rose gradually to 54 degrees centigrade, whereafter the resulting mixture was cooled to 22 degrees centigrade and filtered. The white solid thus obtained was washed twice with a total of 500 milliliters of n-hexane and refiltered. The aqueous layer of the filtrate contained no insoluble matter, and the hexane layer, on evaporation, left a yellow liquid residue weighing only seven grams. The white solid was recrystallized from carbon tetrachloride, placed in an oven at 110 degrees centigrade for ten minutes to remove volatile solvent, and then analyzed without further exposure to ordinary air. It was found to contain 1.75 percent water and to have a neutral equivalent (thymol) of 532. Another sample of the same material was placed in an oven at 108 degrees centigrade for 39 hours and then analyzed. It contained 2.13 percent water and had a neutral equivalent of 538. Another portion of the original solid, which was crystallized from carbon tetrachloride was allowed to stand in contact with ordinary air overnight. It was then analyzed and found to contain 5.82 percent water and to have a neutral equivalent of 559. All of the materials were found to melt within the range 350-357 degrees centigrade and none depressed the melting points of the others. Still another sample of the product was recrystallized from a mixture of acetyl chloride and hexane and kept in a desiccator over phosphorus pentoxide. This material analyzed 0.15 percent water, 71.8 and 71.8 percent chlorine, and had a neutral equivalent of 500. Still another portion has recrystallized from thionyl chloride and kept over phosphorus pentoxide. Analysis of this sample showed 0.19 percent water, 71.26, 71.82 percent chlorine, and a neutral equivalent of 478.

Example 3: Step (b) Hydrolysis of $C_5Cl_6SO_3$ Reaction Product

A 302-gram aliquot of the crude reaction product, prepared as in Example I, was added to one liter of methanol. The temperature rose rapidly to the boiling point of the clear yellow solution. The methanol was evaporated on a steam bath to leave a semi-solid residue. It was cooled, filtered, and dried to yield 136 grams of nearly white solid having a melting point (sealed tube) 351-353 degrees centigrade. Recrystallization from carbon tetrachloride resulted in negligible change in the melting point. No depression of melting point was observed upon mixing this sample and the product obtained by reacting the compound $C_{10}H_2Cl_{12}O_3S$ with activated carbon.

Example 4 Preparation of $C_{10}H_2Cl_{12}O_3S$ from C_5Cl_6-SO_3 Reaction Product and HCl

A 290-gram sample of crude reaction mixture, prepared as in Example 1, was stirred vigorously while anhydrous hydrogen chloride was bubbled into the mixture for four hours at 90-96 degrees centigrade. Solid began to crystallize from the mixture after about 1.5 hours, whereafter the mixture was cooled to 30 degrees centigrade and filtered through a sintered glass funnel. The resulting solid weighed 114 grams. It was recrystallized from 150 milliliters of boiling glacial acetic acid to yield 36 grams of the compound $C_{10}H_2Cl_{12}O_3S$, melting point 150 degrees centigrade (corrected). It did not depress the melting point of an authentic specimen of $C_{10}H_2Cl_{12}O_3S$.

Preparation 1 Preparation of $C_{10}H_2Cl_{12}O_3S$

A mixture of hexachlorocyclopentadiene (4.0 moles, 1092 grams) and chlorosulfonic acid (8.0 moles, 932 grams) was stirred at room temperature while sulfur trioxide (98.5 grams) was added dropwise to the mixture. Upon addition of sulfur trioxide, the reaction mixture became purple in color and then greenish black. As the reaction mixture was heated, with agitation, it became brown and then remained a reddish color until the reaction was completed. The temperature was increased rapidly to ninety degrees centigrade and, after two or three minutes at this temperature, the solid $C_{10}H_2Cl_{12}O_3S$ began to crystallize from the reaction mixture. A slurry of product formed within seven minutes, whereafter the reaction was continued for 2.5 hours at 90 degrees centigrade and then cooled to room temperature and filtered in a sintered glass funnel. The solid was recrystallized directly from glacial acetic acid to produce the compound of the approximate molecular formula $C_{10}H_2Cl_{12}O_3S$ (978 grams) in high purity.

For further description of this starting compound and a method of preparing the same, see U. S. Patent 2 516 404 to McBee and Newcomer, entitled "Reaction Product of Hexachlorocyclopentadiene and Chlorosulfonic Acid", issued 1950 July 25 on an application filed 1948 January 9.

__Example 5__ Preparation of $C_{10}HCl_{10}O_2$ from
$C_{10}H_2Cl_{12}O_3S$ and KOH

During a period of 35 minutes, the compound $C_{10}H_2Cl_{12}O_3S$ (100 grams) was added to a solution of potassium hydroxide (300 grams) in methanol (850 milliliters). The temperature of the vigorous reaction was maintained at 55-65 degrees centigrade by the regulated addition of $C_{10}H_2Cl_{12}O_3S$. The resulting solution was diluted to two liters with water to form a clear solution. Acidification with hydrochloric acid gave a bulky, gelatinous, white precipitate. The solid was filtered, dissolved in acetone, carbon tetrachloride added and the mixture evaporated till crystals began to form. Cooling and filtration gave 64 grams of white crystals. Recrystallization of the solid from carbon tetrachloride-acetone resulted in a melting point (sealed tube) of 350-355 degrees centigrade. It did not depress the melting point of the materials prepared in Examples 2 and 3. This sample was found to contain 64.2 percent chlorine and to have a neutral equivalent (thymolphthalein endpoint) of 588.

__Example 6__ Preparation of $C_{10}HCl_{10}O_2$ from
$C_{10}H_2Cl_{12}O_3S$ and CH_3OH

A solution of the compound $C_{10}H_2Cl_{12}O_3S$ (50 grams) and methanol (1700 milliliters) was refluxed for six hours and then solvent evaporated to the appearance of solid. Four hundred milliliters of carbon tetrachloride was then added and the evaporation repeated. Upon cooling and filtering, 24 grams of white solid was obtained. It was found to contain

67.3 percent chlorine, to have a neutral equivalent of 532, and to melt at 339-359 degrees centigrade. It did not depress the melting point of the hydrated or anhydrous $C_{10}HCl_{10}O_2$ prepared in the previous Examples. Hence, the product of this reaction appears to be a hydrate of the compound of approximate formula $C_{10}HCl_{10}O_2$.

Example 7 Preparation of $C_{10}HCl_{10}O_2$ from
$C_{10}H_2Cl_{12}O_3S$ and KI

A mixture of the compound $C_{10}H_2Cl_{12}O_3S$ (40 grams), potassium iodide (45 grams), and acetone (350 milliliters) was refluxed for 0.5 hour. The mixture remained light in color for a few minutes and then rapidly turned dark. The reaction mixture was diluted to two liters with water, filtered, and "dried" by exposure to air. Recrystallization from a mixture of carbon tetrachloride and acetone gave thirty grams of a solid having 64.2 percent chlorine and melting at 350-352 degrees centigrade. The product does not depress the melting point of previously prepared samples of $C_{10}HCl_{10}O_2$, hydrated as well as anhydrous.

Example 8 Preparation of $C_{10}HCl_{10}O_2$ from
$C_{10}H_2Cl_{12}O_3S$ and Activated Carbon

A mixture of the compound $C_{10}H_2Cl_{12}O_3S$ (627 grams), activated carbon (60 grams), and toluene (1200 grams) was refluxed for seven hours. The hot mixture was filtered and the filtrate evaporated to 500 milliliters. Filtration gave 390 grams of white solid which was "dried" by exposure to air for three days. It was found to have a neutral equivalent of 568 and to melt at 350-357 degrees centigrade (sealed tube). A sample of the produce was dried in an oven at 110 degrees centigrade to constant weight. This material analyzed 70.2 percent chlorine and had a neutral equivalent of 526. The compound $C_{10}HCl_{10}O_2$ has a theoretical analysis of 69.8 percent chlorine and a neutral equivalent of 507. During the drying operation, the sample lost 7.75 percent of its weight. A Karl Fisher water determination of the material exposed to air gave 6.67 percent water. After the oven-drying operation, the sample was exposed to ordinary air until the gain in weight had become

Plaintiff's
Exhibit 1

constant. The sample gained 10.5 percent of water (weight difference) or 10.08 percent of water (potassium fluoride) and the hydrate had a neutral equivalent of 573.

A sample of the original product (neutral equivalent 518) was recrystallized from acetyl chloride-hexane and kept in a vacuum desiccator over sulfuric acid. It analyzed 70.9 percent chlorine and had a neutral equivalent of 513. Upon exposure of the material to air, it gained 11.57 percent of water (weight difference) or 11.2 percent of water (potassium fluoride). The hydrated form has a neutral equivalent of 572.

All of the products prepared in this manner melt (corrected, sealed tube) in the range of 350-357 degrees centigrade and give no depression of melting point when mixed melting points are taken. Further, the materials did not depress the melting point of previously prepared samples of the new material.

During reaction of $C_{10}H_2Cl_{12}O_3S$ with activated carbon, nearly all of the sulfur is liberated as sulfur dioxide. Thus, 0.9 mole of sulfur dioxide was collected from reaction of activated carbon with one mole of $C_{10}H_2Cl_{12}O_3S$ and identified by analysis.

Example 9 Preparation of $C_{10}HCl_{10}O_2$ Potassium Salt

A small sample of the compound $C_{10}H_2Cl_{12}O_3S$ was treated with alcoholic potassium hydroxide as previously described. The resulting alkaline solution was evaporated to precipitate a white solid. The solid was filtered and recrystallized from acetone to form the potassium salt of the acid $C_{10}HCl_{10}O_2$. It was found to contain 62.1 percent chlorine. Theoretical analysis of the salt $C_{10}Cl_{10}O_2K$ is 65 percent chlorine.

Example 10 Preparation of $C_{10}HCl_{10}O_2$Pyridinium Salt

Pyridine (120 grams) was added dropwise to a solution of the compound $C_{10}H_2Cl_{12}O_3S$ (314 grams) in 1050 milliliters of benzene. The resulting mixture was filtered from insoluble material and the benzene solution evaporated to give a white, crystalline solid. Recrystallization from benzene gave a melting point of 143-143 degrees centigrade (sealed tube). The product was found to contain 60.3 percent chlorine, the theoretical for the pyridinium salt of $C_{10}HCl_{10}O_2$ being 60.4 percent chlorine.

Example 11 Preparation of $C_{10}HCl_{10}O_2$ Pyridinium Salt

A small sample of the acid $C_{10}HCl_{10}O_2$ was dissolved in benzene and an excess of pyridine added thereto. Evaporation and crystallization gave white needles having a melting point of 142-145 degrees centigrade, which did not depress the melting point of the product obtained by reacting the compound $C_{10}H_2Cl_{12}O_3S$ with pyridine.

Example 12 Preparation of $C_{10}HCl_{10}O_2$ Diethylammonium Salt

Diethylamine (20 grams) was added dropwise to a solution of the compound $C_{10}HCl_{10}O_2$ (7 grams) dissolved in 125 milliliters methanol. The resulting solution was distilled with water to precipitate the product, which was filtered and crystallized from carbon tetrachloride. The white needles (6 grams) decompose gradually above 150 degrees centigrade and contain 60.6, 60.2 percent chlorine. Theoretical analysis for the diethylammonium salt of the acid $C_{10}HCl_{10}O_2$, having the formula $C_{14}H_{12}Cl_{10}O_2N$, is 61.1 percent chlorine.

Example 13 Preparation of $C_{10}HCl_{10}O_2$ Diethylammonium Salt

Diethylamine was added dropwise to a solution of the compound $C_{10}H_2Cl_{12}O_3S$ (15 grams) in 125 milliliters of methanol. The resulting solution was diluted with water to precipitate the product, which was filtered and crystallized from carbon tetrachloride. The white needles thus obtained (9 grams) were found to contain 61.3, 61.5 percent chlorine. Theoretical analysis for the diethylammonium salt of the acid $C_{10}HCl_{10}O_2$ is 61.1 percent chlorine. The product has the same solubilities as does the product obtained by reacting diethylamine with the acid $C_{10}HCl_{10}O_2$.

Example 14 Preparation of $C_{10}HCl_{10}O_2$ Phenylhydrazinium Salt

The compound $C_{10}HCl_{10}O_2$ (25 grams) was dissolved in 400 milliliters of methanol. Water was then added until the solution became milky. A little methanol was added to give a clear solution, whereafter

Plaintiff's
Exhibit 1

25 grams of phenylhydrazine was added. Upon heating for five minutes on a steam bath and cooling, there was obtained 32 grams of faintly yellow needles, melting point 159.5-161.1 degrees centigrade; 56.3 percent chlorine. The theoretical analysis for the phenylhydrazinium salt of $C_{10}HCl_{10}O_2$ is 57.6 percent chlorine. A sample of the salt was dissolved quickly in ethyl alcohol at room temperature and acidified with aqueous hydrochloric acid. The precipitate was filtered, washed with water, and dried in an oven at 110 degrees centigrade. It was found to melt at 341-348-357 degrees centigrade and did not depress the melting point of the original $C_{10}HCl_{10}O_2$.

Example 15 Stability of $C_{10}HCl_{10}O_2$

A mixture of forty grams of the compound $C_{10}HCl_{10}O_2$, ten grams of potassium hydroxide, and 200 grams of potassium permangante, in five liters of water, was refluxed for five hours. The reaction mixture was then worked up to give 36 grams of unreacted starting material, as shown by melting point and mixed melting point determinations.

It is to be understood that the invention is not to be limited to the exact details of operation or exact compounds shown and described, as obvious modifications and equivalents will be apparent to one skilled in the art to which the invention pertains, and the invention is therefore to be limited only by the scope of the appended claims.

We Claim:

- 1 -

A new composition of matter selected from the group consisting of (a) A compound of the approximate molecular formula $C_{10}HCl_{10}O_2$, characterized by having a corrected melting point in a sealed tube of about 355 degrees centigrade and in that it contains about seventy percent chlorine by weight when in an anhydrous state, being substantially identical with the product formed by treating, with water, the reaction product of hexachlorocyclopentadiene and sulfur trioxide, and then dehydrating the product, (b) hydrates thereof, and (c) salts thereof.

-2-

A compound of the approximate molecular formula $C_{10}HCl_{10}O_2$, characterized by having a corrected melting point in a sealed tube of about 355 degrees centigrade and in that it contains about seventy percent chlorine by weight when in an anhydrous state, being substantially identical with the product formed by treating, with water, the reaction product of hexachlorocyclopentadiene and sulfur trioxide, and then dehydrating the product.

-3-

A salt of the compound defined in Claim 2.

-4-

A hydrate of the compound defined in Claim 2.

OATH, POWER OF ATTORNEY, AND PETITION

Being duly sworn, We, EARL T. McBEE and JACK S. NEWCOMER depose and say:

THAT We are citizens of the United States of America residing in the City of West Lafayette, County of Tippecanoe, State of Indiana and City of Niagara Falls, County of Niagara, State of New York, respectively.

THAT we have read the foregoing specification and claims and We verily believe We are the original, first and joint inventors of the invention or discovery in

COMPOSITION OF MATTER

described and claimed therein;

THAT We do not know and do not believe that this invention was ever known or used before our invention or discovery thereof, or patented or described in any printed publication in any country before our invention or discovery thereof, or more than one year prior to this application, or in public use or on sale in the United States for more than one year prior to this application;

THAT this invention or discovery has not been patented in any country foreign to the United States on an application filed by us or our legal representatives or assigns more than twelve months before this application; and

Plaintiff's
Exhibit 1

THAT no application for patent on this invention or discovery has been filed by us or our representatives or assigns in any country foreign to the United States.

AND We hereby appoint Dean Laurence (Reg. No. 14 153) Saint Johns, Michigan, our attorney with full power of substitution and revocation, to prosecute this application and to transact all business in the Patent Office connected therewith.

WHEREFORE We pray that Letters Patent be granted to us for the invention or discovery described and claimed in the foregoing specification and claims, and we hereby subscribe our names to the foregoing specification and claims, oath, power of attorney, and this petition.

Signed by me this 17 day of July, 1951.

Inventor: /s/ Earl T. McBee

Post Office Address: 420 Forest Hill Drive
West Lafayette, Indiana

State of Indiana)
: ss.
County of Tippecanoe)

Before me personally appeared EARL T. McBEE, to me known to be the person described in the above application for patent, who signed the foregoing instrument in my presence, and made oath before me to the allegations set forth therein as being under oath, on the day and year aforesaid.

/s/ Jane T. Gaines (McBee)
Notary Public

My Commission Expires: August 9, 1953

Signed by me this 6 day of July, 1951.

Inventor: /s/ Jack S. Newcomer

Post Office Address: Hooker Electrochemical Company
Niagara Falls, New York

State of New York)
: ss.
County of Niagara)

Before me personally appeared JACK S. NEWCOMER, to me known to be the person described in the above application for patent, who signed the foregoing instrument in my presence, and made oath before me to the allegations set forth therein as being under oath, on the day and year aforesaid.

/s/ Alice Young
Notary Public

My Commission Expires: March 30, 1952

Paper No. 2

[Mailed, March 21, 1952, Patent Examining Division...31]

DEPARTMENT OF COMMERCE
UNITED STATES PATENT OFFICE
Washington

Please find below a communication from
the EXAMINER in charge of this application.

/s/ John A. Marzall
Commissioner of Patents.

Applicant:	Earl T. McBee et al.
Ser. No.	238,186
Filed	July 23, 1951
For	COMPOSITION OF MATTER

Dean Laurence
Box 151
One Clinton Avenue
St. Johns, Michigan

This application has been examined.

References cited to show the state of the Art:

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McBee et al	2,552,568	May 15, 1951	260-586

Claims 1-4 are rejected as indefinite and as not properly defining the invention. Since the structure of the product is not known, it must be defined by the method of making it. Ex parte Fessenmier 1922 C. D. 18. The product may not be claimed as being "substantially

Plaintiff's
Exhibit 1

identical" with the product of a particular process. Ex parte Lichty 64 USPQ 430. See also Ex parte Appeal No. 47,564, Case No. 121, 26 JPOS 288.

ESC/els Examiner

IN THE UNITED STATES PATENT OFFICE

Division : 31
Applicant : Earl T. McBee et al.
Serial : 238 186
Filed : 1951 July 23
For : COMPOSITION OF MATTER

Hon. Commissioner of Patents
Washington 25, D. C.

Sir:

AMENDMENT UNDER RULE 115

In response to the Office action mailed 1952 March 21, please amend the above identified application as follows:

IN THE CLAIMS:

Cancel Claims 1-4, inclusive, and rewrite as:

-5-

A material selected from the group consisting of (A) a composition of matter which has the following characteristics: it is a white crystalline solid having a sealed tube corrected melting point of about 355 degrees centigrade, it behaves as a weak organic acid, is very soluble in polar organic solvents, is substantially insoluble in non-polar organic solvents, and has the approximate molecular formula $C_{10}HCl_{10}O_2$; (B) hydrates of (A); and, (C) salts of (A).

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A composition of matter which has the following characteristics: it is a white chrystalline solid having a sealed tube corrected melting point of about 355 degrees centigrade, it behaves as a weak organic acid, is very soluble in polar organic solvents, substantially insoluble in non-polar organic solvents, and has the approximate molecular formula $C_{10}HCl_{10}O_2$.

-7-

A salt of the compound defined in Claim 6.

-8-

A hydrate of the compound defined in Claim 6.

REMARKS

The claims have been rewritten to eliminate the rejection on the ground of indefiniteness and to remove the objectionable "substantially identical" language.

Issue is taken on the statement by the Examiner that "Since the structure of the product is not known, it must be defined by the method of making it.", citing Ex parte Fesenmeier 1922 CD 18. As stated by the Commissioner at page 21, the question in Fesenmeier was:

> "The question presented for review is whether the instant case is one to which the rule against claims of this character should be applied, or whether it is one forming a just exception, one where the product is new and involves inventive concept in its production, and yet can not be defined save by stating the steps necessary to produce it."

Nothing is observed in the opinion which would support the above-quoted statement of the Examiner.

The claims submitted are in the form approved in Mauer v. Dickerson et al. 113 Fed. 870 by the Court of Appeals for the Third Circuit. No authority is known for the proposition that, lacking a structural formula, but given a molecular or empirical formula and other physical properties, a composition of matter must be defined by the method of making it. It is noted the Commissioner in Ex Parte Fesenmeier, at page 21, stated that the characteristics of a product,

Plaintiff's
Exhibit 1

whereby it may be identified, did not have to be such that the general public could, by a mere casual inspection, determine the novel features of the product. The Commissioner further stated that the product might have to be tested chemically to determine its characteristics "but if these are so set forth that they may be determined in one of these ways it is sufficient". Here, it is considered that sufficient properties are given in the claims and in the specification whereby a chemist could determine the boundaries of the claimed invention and ascertain readily the issue of infringement, should such need arise. This meets the requirements of the law.

It is submitted the rewritten claims are in proper form and overcome the rejection of the above-identified Office action and allowance is respectfully solicited.

Respectfully,

/s/ Dean Laurence
Attorney

Saint Johns, Michigan

1952 September 9
4CO:DL:kw
PRF 4.1-517

IN THE UNITED STATES PATENT OFFICE

Division : 31

Applicant : Earl T. McBee et al.

Serial : 238 186

Filed : 1951 July 23

For : COMPOSITION OF MATTER

Hon. Commissioner of Patents
Washington 25, D. C.

Sir:

SUPPLEMENTAL AMENDMENT UNDER RULE 115

In supplemental response to the Office action mailed 1952

March 21, and the amendment under Rule 115 mailed to the Patent Office approximately 1952 September 16, please amend the above-identified application for patent as follows:

In the Specification:

Page 1, line 3, cancel the formula and insert --- $C_{10}Cl_{10}O$ ---.
line 4, cancel "elemental composition and" and insert
--- molecular ---.
line 5, cancel "are" and insert --- is ---.
line 26, cancel the formula and insert
--- $C_{10}Cl_{10}O$ ---

Page 3, line 1, after "mineral" insert --- acid ---.
lines 3, 4 and 5, cancel the lines.
line 6, cancel "holic caustic solution."

Page 4, line 3, cancel the formula and insert --- $C_{10}Cl_{10}O$ ---.
line 9, after "solids" insert --- usually ---.
line 10, cancel "The anhydrous form has a tendency to".
line 11, cancel "acquire a marked electrostatic charge.";
before the word "compound" insert --- monohydrate of the ---.
line 12, cancel the formula and insert --- $C_{10}Cl_{10}O$ ---.
line 13, cancel the comma and insert a period; cancel "but a".
line 14, cancel "definite end-point with thymophthalein."

Page 6, cancel the formula in the heading of the table and insert
--- $C_{10}Cl_{10}O$ ---.

Page 7, line 1, cancel the formula and insert --- $C_{10}Cl_{10}O$ ---.
line 4, cancel the formula and insert --- $C_{10}Cl_{10}O$ ---.
line 7, cancel the formula and insert --- $C_{10}Cl_{10}O$ ---.
line 12, cancel the formula and insert --- $C_{10}Cl_{10}O$ ---.
line 27, cancel the formula and insert --- $C_{10}Cl_{10}O$ ---.
line 30, cancel the formula and insert --- $C_{10}Cl_{10}O$ ---.

Page 8, line 3, cancel the second formula and insert
--- $C_{10}Cl_{10}O$ ---.
line 7, cancel the formula and insert --- $C_{10}Cl_{10}O$ ---.

Plaintiff's
Exhibit 1

line 15, cancel the first formula and insert

--- $C_{10}Cl_{10}O$ ---.

Page 12, line 4, cancel the first formula and insert

--- $C_{10}Cl_{10}O$ hydrate ---.

line 22, cancel the first formula and insert

--- $C_{10}Cl_{10}O$ ---.

Page 13, line 1, cancel the formula and insert --- $C_{10}Cl_{10}O$ ---.

line 4, cancel the formula and insert --- $C_{10}Cl_{10}O$ ---.

line 5, cancel the formula and insert --- $C_{10}Cl_{10}O$ ---.

line 15, cancel the formula and insert --- $C_{10}Cl_{10}O$ ---.

line 17, cancel the formula and insert --- $C_{10}Cl_{10}O$ ---.

Page 14, line 25, cancel the formula and insert --- $C_{10}Cl_{10}O$ ---.

Page 15, line 3, cancel the formula and insert --- $C_{10}Cl_{10}O$ ---.

line 13, cancel the formula and insert --- $C_{10}Cl_{10}O$ ---.

line 14, cancel the formula and insert --- $C_{10}Cl_{10}O$ ---.

line 20, cancel the formula and insert --- $C_{10}Cl_{10}O$ ---.

line 22, cancel the formula and insert --- $C_{10}Cl_{10}O$ ---.

Page 16, line 1, cancel the formula and insert --- $C_{10}Cl_{10}O$ ---.

line 11, cancel "the acid"; cancel the formula and insert

--- $C_{10}Cl_{10}O$ ---.

line 12, cancel the formula and insert --- $C_{10}Cl_{10}O$ ---.

line 13, cancel the formula and insert --- $C_{10}Cl_{10}O$ ---.

line 27, cancel the formula and insert --- $C_{10}Cl_{10}O$ ---.

line 28, cancel the formula and insert --- $C_{10}Cl_{10}O$ ---.

line 29, cancel the formula and insert --- $C_{10}Cl_{10}O$ ---.

In the Claims:

Claim 5, line 8, cancel the formula and insert --- $C_{10}Cl_{10}O$ ---.

Claim 6, line 7, cancel the formula and insert --- $C_{10}Cl_{10}O$ ---.

REMARKS

The above amendments to the specification and claims bring this application in line with the technical facts which have been developed. It has been found by exhaustive research procedures that the original formula given for the compound herein described and claimed was, in fact, incorrect. The formula inserted in the specification and claims, i.e., $C_{10}Cl_{10}O$, is the exact formula of the compound prepared by the method of and having the properties defined in the specification. Authority for this amendment is found in Ex parte Harris 43 USPQ 125, 1939 Board of Appeals decision, Appeal 28 470. In that case, it was stated that inherent properties of a material may be introduced into the specification by way of amendment and that such amendment does not constitute the addition of new matter thereto. Here, the formula of the material is certainly an inherent property and since it is the same material which is described very completely in some seventeen pages of specification, it is submitted that such additions and amendments as above proposed do not constitute the addition of new matter.

The procedures whereby the substance claimed can be prepared remain the same and the physical characteristics whereby it can be identified, such as its melting point, solubility, et cetera, remain the same. Had the formula been omitted entirely it could have been inserted on the authority of Ex parte Harman 86 Q 487 and Ex parte Harris, supra, and correction is therefore proper on the same authority and the file of Patent 2 574 519.

It is regretted that the errors which are corrected by this amendment have occurred. However, this material has a molecular weight in excess of approximately 400, and exact determination thereof is very difficult. It is respectfully requested that this amendment be entered and that the prosecution of this application be continued with the amended specification and claims.

Respectfully,

/s/ Dean Laurence
Attorney

Saint Johns, Michigan
1952 September 30
5CO:JBV:kw:DL
PRF 4.1-517

Plaintiff's
Exhibit 1

[Mailed June 17, 1953, Pat. Div. 6]

Paper No. 5

DEPARTMENT OF COMMERCE
UNITED STATES PATENT OFFICE
Washington

XIII Levinson: cln

Dean Laurence
Box 151
One Clinton Ave.
St. Johns, Mich.

Applicant: Earl T. McBee et al
Ser. No. 238,186
Filed July 23, 1951
For COMPOSITION OF MATTER

Please find below a communication from the EXAMINER in charge of this application.

/s/ Robert C. Watson

Commissioner of Patents

Responsive to amendments filed Sept. 18, 1952 and Oct. 2, 1952.

Claims 5-8, inclusive, remain in the case.

The empirical formulae appearing on page 14, line 30; page 15, lines 11, 28 and 29; page 16, lines 8, 21, and 28 of the specification are queried. These formulae do not appear to agree with the amended formulae elsewhere in the specification and claims.

Claims 5-8, inclusive, are rejected as indefinite and improper product claims. Since the structure of the product is not known, it must be defined by its method of manufacture. Ex parte Fessenmier, 1922 C.D. 18; Ex parte Serini et al, 68 USPQ 282.

Since an issue appears to have been reached, the rejection of claims 5-8, inclusive, is made FINAL.

Examiner

Plaintiff's
Exhibit 1

IN THE UNITED STATES PATENT OFFICE

Division : 6
Applicant : Earl T McBee et al
Serial : 238 186
Filed : 1951 July 23
For : COMPOSITION OF MATTER

Hon Commissioner of Patents
Washington 25 D C

AMENDMENT UNDER RULE 116

Sir:

In response to the Office Action mailed 1953 June 17, a Final Rejection, please amend as follows:

In the Claims:

Claim 5, line 7: cancel "and".
line 8: after the formula and before the semicolon insert ---and which may be prepared by reacting $C_{10}H_2Cl_{12}O_3S$ with active hydrogen atoms and subsequent dehydration ---.
Claim 6, line 6: cancel "and"
line 7: after the formula and before the period insert ---which may be prepared by reacting $C_{10}H_2Cl_{12}O_3S$ with active hydrogen atoms and subsequent dehydration---.

Please add the following new claims:

A composition of matter which has the following characteristics: it is a white crystalline solid having a sealed tube corrected melting point of about 355 degrees Centigrade, it behaves as a weak organic acid, is very soluble in polar organic solvents, substantially insoluble in non-polar organic solvents and has the approximate molecular formula $C_{10}Cl_{10}O$ and is prepared by the reaction of $C_{10}H_2Cl_{12}O_3S$

with a basic substance, subsequent treatment with an acid, and dehydration.

-10-

A composition of matter which has the following characteristics: it is a white crystalline solid having a sealed tube corrected melting point of about 355 degrees Centigrade, it behaves as a weak organic acid, is very soluble in polar organic solvents, substantially insoluble in non-polar organic solvents, has the approximate molecular formula $C_{10}Cl_{10}O$ and is prepared by the reaction of $C_{10}H_2Cl_{12}O_3S$ with an inorganic iodide and subsequent dehydration.

-11-

A composition of matter which has the following characteristics: it is a white crystalline solid having a sealed tube corrected melting point of about 355 degrees Centigrade, it behaves as a weak organic acid, is very soluble in polar organic solvents, substantially insoluble in non-polar organic solvents and has the approximate molecular formula $C_{10}Cl_{10}O$ and is prepared by the reaction of $C_{10}H_2Cl_{12}O_3S$ with activated carbon and subsequent dehydration.

-12-

A composition of matter which has the following characteristics: it is a white crystalline solid having a sealed tube corrected melting point of about 355 degrees Centigrade, it behaves as a weak organic acid, is very soluble in polar organic solvents, substantially insoluble in non-polar organic solvents, has the approximate molecular formula $C_{10}Cl_{10}O$ and is prepared by the reaction of $C_{10}H_2Cl_{12}O_3S$ with active hydrogen atoms and subsequent dehydration.

REMARKS

It is desired to thank the Examiner for the interview afforded applicants' attorney recently. The discussion during the interview included the following:

With regard to the empirical formulae appearing at pages 14, 15 and 16, which were not amended, it is to be noted that each relates to a hydrated material and not to the dehydrated material of the amendments. It is desired, however, to thank the Examiner for his careful perusal of the specification and the pointing out of the apparent discrepancy.

It will be noted that all of the claims have been amended to insert the method of preparation whereby the compound of the claims may be prepared. In certain instances, the claims have been directly limited to a definite method of preparation, but, in view of the facts to be hereinafter discussed, it is submitted that it is not necessary that all of the claims be limited to the particular method of preparation.

It is to be noted that the compound of the present invention has outstanding utility in the field of desiccation, as well as having utility in the field of insecticides. The compound of the present invention may be prepared in at least four different manners which are discussed in the application, including as a starting material in each case the treatment of $C_{10}H_2Cl_{12}O_3S$, which is disclosed in United States Patent 2 516 404 issued 1950 July 25. Thus, the reaction of this material with a basic substance and subsequent treatment with acid, an inorganic iodide, with activated carbon, or with active hydrogen atoms will result in the production of a compound having the molecular formula $C_{10}Cl_{10}O$. It is to be noted that the problem of dehydration was very difficult in the matter of the compound of this invention, since it tends to form hydrates with any water which might be present. Dehydration has been accomplished only with difficulty. Necessarily, therefore, the above reactions will result in a hydrated material and only by a dehydration procedure may the non-hydrated material be prepared.

Ex parte Fessenmeir, 1922 C.D. 18 has been noted. However, this case is not applicable to the facts of the present application. In Ex parte Fessenmeir the claims related to a product which could not be defined by a structural or molecular formula, nor could it be definitely characterized such as the compound of the present invention has been characterized. The question in that case was whether it was proper to allow claims to a material which has only been defined by a method of production. It was held that the exception to the general rule in not allowing this type of claim should be allowed in the Fessenmeir case.

In the instant application, however, such facts are not present. The compound of the present invention has been definitely characterized by formula, by solubilities, and by melting point. As such, the only thing which remains to accomplish would be the structural formula and no question would be presented. However, in the difficult field of chemistry which this encompasses, the accomplishment of the resolution of structural formula in a laboratory has been found to be extremely difficult if not impossible. In fact, at the present time, the structural formula has not yet been definitely determined.

However, the molecular formula, the melting point, et cetera, have been definitely determined and are set out properly in the claims. There is no question as to what compound is covered, nor would there be any question if the compound was produced by a method other than those disclosed in the specification as to whether the compound of the present application was in fact being infringed. There has been a substantial contribution to the art, and consideration of these facts is respectfully requested.

In Ex parte Serini and Steinbrook, 68 USPQ 282, the question was what compound was prepared. There were in fact two possible compounds prepared by the procedure and it was held that since there were two possibilities and since the art showed at least one of the possibilities, the method of production must necessarily limit the product claims. However, here again, these facts are substantially different from the facts which are presented by the instant application. In the instant application, a definitely characterized compound is being claimed, having a definite structure, having a definite melting point, and having definite chemical and physical properties and there is no art which is pertinent to the compound of the present invention.

The claims have been generally rewritten and there is no question of what compound is being claimed. As such, it is submitted that the

Serini et al decision is not pertinent to the facts of the present application.

The claims of the present application being definite and describing only a specific compound, its hydrates and salts, it is submitted that they are properly allowable.

Reconsideration and allowance is respectfully requested.

In the event that this amendment does not place this application is condition for allowance, entry for purposes of appeal is respectfully requested.

Respectfully

/s/ Dean Laurence
Attorney

Saint Johns, Michigan
1953 September 16
JBV je 4C

[Mailed September 30, 1953, Pat. Div. 6]

Paper No. 7

DEPARTMENT OF COMMERCE
UNITED STATES PATENT OFFICE
Washington

XIII-Levinson:

Dean Laurence
Box 151
One Clinton Ave.
St. Johns, Mich.

Applicant: Early T. McBee et al.
Ser. No. 238,186
Filed July 23, 1951
For COMPOSITION OF MATTER

Please find below a communication from the EXAMINER in charge of this application.

/s/ Robert C. Watson
Commissioner of Patents.

Responsive to amendment filed September 21, 1953 after final rejection.

The amendment has not been entered since it does not place the case in condition for allowance for the reasons discussed below. The amendment will not be entered for purpose of appeal since it raises new issues.

The empirical formulae appearing on page 14, line 30; page 15, lines 2, 11, 28, and 29; page 16, lines 8, 21, and 28 of the specification are again queried. The explanation that these formulae refer to a hydrated $C_{10}Cl_{10}O$ is considered unsatisfactory since there is still a deficiency of one hydrogen atom.

The proposed amendments cannot be entered since they render the present claims indefinite in the recitation of "may be prepared". Since the structural formula of the compound is unknown, the products must be recited by their process of manufacture which in turn must be positively set forth in the claims. Furthermore, the processes recited in the claims are too broad. In all the examples, heating is necessary. The only support for "basic substance" is potassium hydroxide which is insufficient. The only support for an "inorganic iodide" is potassium iodide which is insufficient. The only support for "active hydrogen atoms" is methanol which is obviously insufficient.

The following claim would probably be allowable:

> A composition of matter which has the following characteristics: it is a white crystalline solid having a sealed tube corrected melting point of about 355 degrees Centigrade, it behaves as a weak organic acid, is very soluble in polar organic solvents, substantially insoluble in non-polar organic solvents and has the approximate formula $C_{10}Cl_{10}O$ and is prepared by the heating of $C_{10}H_2Cl_{12}O_3S$ with a member of the group consisting of potassium hydroxide, methanol, potassium iodide, and activated carbon and subsequent dehydration.

This case will be abandoned unless placed in condition for allowance or appeal is taken on or before December 17, 1953.

Examiner

Plaintiff's
Exhibit 1

IN THE UNITED STATES PATENT OFFICE

Division : 6
Applicant : Earl T McBee et al
Serial : 238 186
Filed : 1951 July 23
For : COMPOSITION OF MATTER

Hon Commissioner of Patents
Washington 25 D C

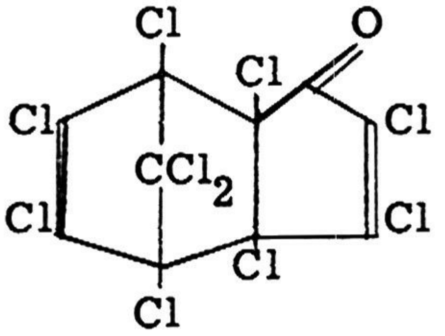
AMENDMENT UNDER RULE 116

Sir:

In response to the Office Action mailed 1953 June 17, and the Supplemental Action mailed 1953 September 30, please amend as follows:

In the Specification:

Page 1, line 3: cancel the period and "While the" and insert ---, and the following structural formula:



lines 4, 5, 6, 7, and 8: cancel the lines.

Page 2, line 26: cancel "any"

Page 7, line 2: cancel "The data presented in".

lines 3, 4, and 5: cancel the lines.

Page 9, line 3: cancel "Upon cooling the pot residue to room".

line 4: cancel "temperature, the product solidified as a tan solid".

Page 14, line 25: cancel the formula "$C_{10}HCl_{10}O_2$" and insert therefor the formula ---$C_{10}H_2Cl_{10}O_2$---.

line 30: cancel the formula "$C_{10}HCl_{10}O_2$" and insert therefor the formula ---$C_{10}H_2Cl_{10}O_2$---.

Plaintiff's
Exhibit 1

Page 15, line 2: cancel the formula "$C_{10}Cl_{10}O_2K$" and insert therefor the formula ---$C_{10}Cl_{10}O_2KH$---.

line 3: cancel the formula "$C_{10}HCl_{10}O_2$" and insert therefor the formula ---$C_{10}H_2Cl_{10}O_2$---.

line 11: cancel the formula "$C_{10}HCl_{10}O_2$" and insert therefor the formula ---$C_{10}H_2Cl_{10}O_2$---.

line 13: cancel the formula "$C_{10}HCl_{10}O_2$" and insert therefor the formula ---$C_{10}H_2Cl_{10}O_2$---.

line 14: Cancel the formula "$C_{10}HCl_{10}O_2$" and insert therefor the formula ---$C_{10}H_2Cl_{10}O_2$---.

line 20: cancel the formula "$C_{10}HCl_{10}O_2$" and insert therefor the formula ---$C_{10}H_2Cl_{10}O_2$---.

line 22: cancel the formula "$C_{10}HCl_{10}O_2$" and insert therefor the formula ---$C_{10}H_2Cl_{10}O_2$---.

line 28: cancel the formula "$C_{10}HCl_{10}O_2$" and insert therefor the formula ---$C_{10}H_2Cl_{10}O_2$---.

line 29: cancel the formula "$C_{10}HCl_{10}O_2$" and insert therefor the formula ---$C_{10}H_2Cl_{10}O_2$---.

Page 16, line 1: cancel the formula "$C_{10}HCl_{10}O_2$" and insert therefor the formula ---$C_{10}H_2Cl_{10}O_2$---.

line 8: cancel the formula "$C_{10}HCl_{10}O_2$" and insert therefor the formula ---$C_{10}H_2Cl_{10}O_2$---.

line 11: cancel the formula "$C_{10}HCl_{10}O_2$" and insert therefor the formula ---$C_{10}H_2Cl_{10}O_2$---.

line 12: cancel the formula "$C_{10}HCl_{10}O_2$" and insert therefor the formula ---$C_{10}H_2Cl_{10}O_2$---.

line 13: cancel the formula "$C_{10}HCl_{10}O_2$" and insert therefor the formula ---$C_{10}H_2Cl_{10}O_2$---.

line 21: cancel the formula "$C_{10}HCl_{10}O_2$" and insert therefor the formula ---$C_{10}H_2Cl_{10}O_2$---.

line 27: cancel the formula "$C_{10}HCl_{10}O_2$" and insert therefor the formula ---$C_{10}H_2Cl_{10}O_2$---.

Plaintiff's
Exhibit 1

line 28: cancel the formula "$C_{10}HCl_{10}O_2$" and insert therefor the formula ---$C_{10}H_2Cl_{10}O_2$---.

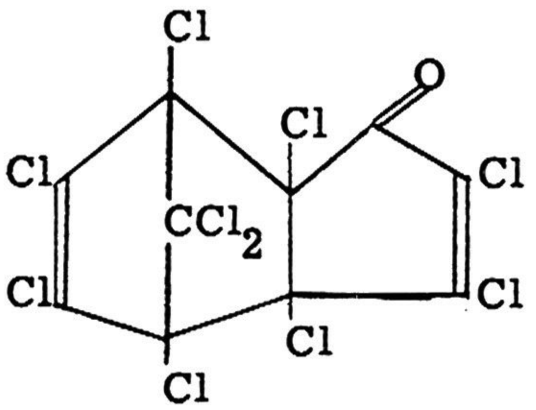
line 29: cancel the formula "$C_{10}HCl_{10}O_2$" and insert therefor the formula ---$C_{10}H_2Cl_{10}O_2$---.

In the Claims:

Cancel all of the claims presently pending in this application and add the following new claims:

-9-

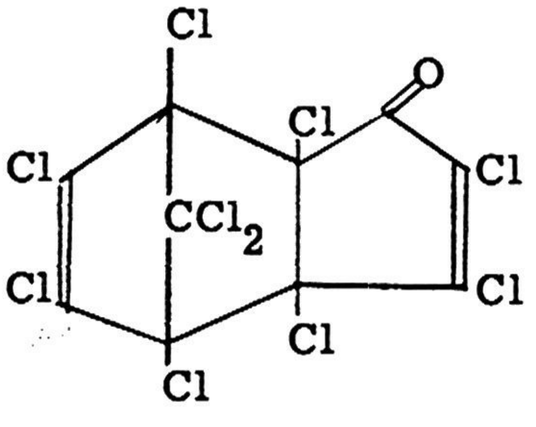
A compound selected from the group consisting of (a) a compound having the formula:



characterized by having a corrected melting point in a sealed tube of about 355 degrees Centigrade, (b) hydrates of this compound and (c) salts thereof.

-10-

A compound having the formula



Plaintiff's
Exhibit 1

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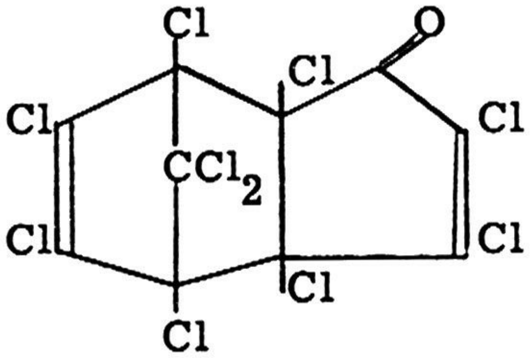
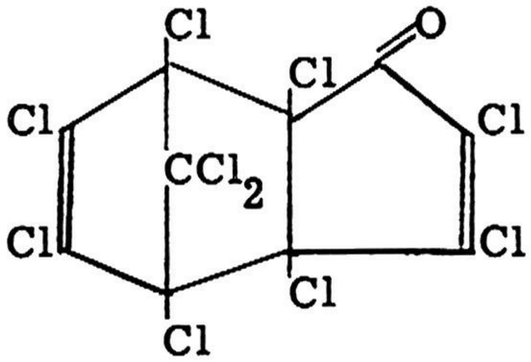
A salt of a compound having the formula:



-12-

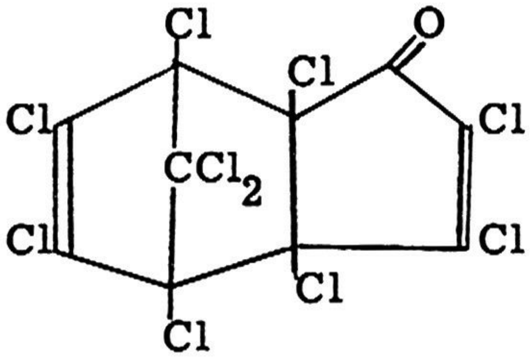
A hydrate of a compound having the formula:



REMARKS

It is desired to thank the Examiner for the cordial interview recently afforded applicants' attorney. The careful consideration of applicants' problems at that time is gratefully appreciated.

As will be noted from this amendment, the structural formula has now been conclusively determined by the inventors and is as follows:



It is regretted that the structural formula was not known at an earlier time, and that it was impossible to know that the structural formula would be determined. Applicants' attorney has only recently been informed thereof. At the same time, the formula has not been known to the inventors and only very recently did they conclusively determine that it is as above.

Plaintiff's
Exhibit 1

Each of the amendments to the application does not constitute new matter and may properly be entered under the authority of Ex parte Harris, 23 USPQ 125 and Ex parte Harmon 86 USPQ 487. Further authority may be found in Patent file 2 574 519. All of these cases state that the structural formula upon being determined can be inserted in the application. This is being done in this application. It will be noted also that a hydrogen atom has been added to certain of the formulae in the specification. With the structural formula now being known, it may be known definitely that the hydrated material has the addition of an H_2O to the known structural formula. This is believed to clarify the query of the empirical formulae appearing at pages 14, 15 and 16.

The newly-submitted claims are clearly supported in the specification as filed and are clearly allowable over any art which has been cited. It is to be noted that the material is a ketone-type containing ten chlorine atoms therein and two double bonds, and the structural formula has been conclusively identified in the claims. As such, it is submitted that this formula and the claims are clearly allowable and such action is respectfully requested.

It is again to be emphasized that applicants' attorney is extremely regretful of the fact that this information was not previously available since it would have cut down the extended prosecution necessary in this application. However, it was unavailable to applicants' attorney, assignee, their licensees, or to applicants themselves until very recently. Apology therefor is submitted.

It is submitted that this application is now allowable and such action is respectfully requested. In the event that this application is not allowable, it is respectfully requested that this amendment be entered for purposes of appeal.

Respectfully

/s/ Dean Laurence
Attorney

Saint Johns, Michigan
1953 October 8
JBV je 4C

Plaintiff's
Exhibit 1

IN THE UNITED STATES PATENT OFFICE

Applicant: Earl T McBee et al
Serial : 238 186
Filed : 1951 July 23
For : COMPOSITION OF MATTER

Hon Commissioner of Patents
Washington 25 D C

Sir:

Herewith Supplemental Oath under Rule 67 in the above-identified application.

Respectfully

/s/ Dean Laurence
Attorney

* * * * *

SUPPLEMENTAL OATH UNDER RULE 67

EARL T McBEE and JACK S NEWCOMER, whose application for Letters Patent for an improvement in COMPOSITION OF MATTER, Serial 238 186, filed 1951 July 23, being duly sworn, depose and say:

THAT the subject matter of all of the amendments to the specification and claims of their application previously made (including new Claims 9, 10, 11 and 12) was part of their application, was invented before they filed their original application, above-identified, for such invention; and

THAT they do not know and do not believe that the same was ever known or used before their invention or discovery thereof, or patented or described in any printed publication in any country before their invention or discovery thereof, or more than one year prior to their application, or in public use or on sale in the United States for more than one year before the date of this application; and

THAT said invention has not been patented in any foreign country on an application filed by them or their legal representatives or assigns more than twelve months prior to their application in the United States, and has not been abandoned.

/s/ Earl T. McBee

State of Indiana)
: ss
County of Tippecanoe)

Sworn to and subscribed before me this 12th day of October, 1953.

/s/ Phyllis Anne Hebein
Notary Public

My Commission expires: 2-6-57

/s/ Jack S. Newcomer

State of New York)
: ss
County of Niagara)

Sworn to and subscribed before me this 19th day of October, 1953.

/s/ Alice Young
Notary Public

My Commission expires: 3-30-54

Saint Johns, Michigan
1953 October 7
JBV je 4C

[Mailed October 27, 1953, Pat. Div. 31]

DEPARTMENT OF COMMERCE
UNITED STATES PATENT OFFICE
Washington

Paper No. 9

Dean Laurence
Box 151
One Clinton Avenue
St. Johns, Michigan

Applicant: Earl T. McBee et al
Ser. No: 238,186
Filed July 23, 1951
For COMPOSITION OF MATTER

Please find below a communication from the EXAMINER in charge of this application.

/s/ Robert C. Watson

Commissioner of Patents.

Plaintiff's
Exhibit 1

Responsive to amendment filed October 12, 1953.

Additional reference relied on:

Gilbert et al	2,616,928	Nov. 4, 1952 (Filed November 7, 1950)	260-586

(1) The final rejection has been withdrawn and the instant case is being reopened in view of the newly cited art above. Applicants' last amendment has not been entered.

(2) Newly added claim 9 is rejected as unpatentable over Gilbert et al, cited, who disclose a compound whose optical configuration makes it an enantimorph or mirror isomer of applicants' compound. As such the compounds are not considered patentably distinct.

(3) Claim 9 is further rejected as drawn to new matter. The citations relied upon by the applicants in their last correspondence, paper No. 8D, October 12, 1953, are not deemed in point, particularly "Ex parte Harris, 23 U.S.P.Q. 125" which is non-existent. Was Ex parte Harris 43 U.S.P.Q. 125 intended? If so, no relevancy is noted there either. Support for claim 9 is not deemed to be inherent in the original disclosure; this is especially true since applicants have had to amend their original empirical formula in order for it to correspond to the compound they now recite structurally.

(4) Since the art which is newly cited is occasioned by applicants' latest amendment this rejection is made FINAL.

FWG/cjn Examiner

IN THE UNITED STATES PATENT OFFICE

1953 Nov. 13
1ED1: 360:CKE:sjm
PRF 4-1-517

Division : 31
Applicant : Earl T. McBee et al.
Serial : 238,186
Filed : 1951 July 23
For : COMPOSITION OF MATTER

Plaintiff's
Exhibit 1

Honorable Commissioner of Patents
Washington 25, D. C.

LETTER OF INQUIRY

Sir:

Please advise if a Supplemental Oath Under Rule 67 was recently received by the Patent Office in the matter of the above-identified application.

This Supplemental Oath was mailed from our Saint Johns, Michigan office 1953 October 21st.

Respectfully,

/s/ Dean Laurence
Attorney

The supplemental oath under Rule 67, referred to above was received October 23, 1953, and made of record on said date.

/s/ N. Manuelstein
Examiner

IN THE UNITED STATES PATENT OFFICE

Division : 31
Applicant : Earl T. McBee et al
Serial : 238 186
Filed : 1951 July 23
For : COMPOSITION OF MATTER

1954 April 12
FDL:460:DL:cc
PRF 4-1-517

AFFIDAVIT UNDER RULE 131

EARL T McBEE and JACK S. NEWCOMER, being duly sworn, depose and say:

That they are the applicants in the above-identified application;

That they did reduce to practice the invention defined in proposed Claim 13 of said application prior to 1950 November 7 in this country;

That such reduction to practice included the following acts:

A mixture of 1000 grams of chlorosulfonic acid and 1000 grams of hexachlorocyclopentadiene was stirred vigorously for two hours at 90 degrees centigrade. Red crystals began to separate from

Plaintiff's
Exhibit 1

the reaction mixture after 0.5 hour. The thick reaction mixture was cooled to 3 degrees centigrade and maintained at that temperature for 2 hours. The slurry was filtered through a sintered glass funnel to obtain the solid product which was washed with water and then with 400 milliliters of petroleum ether saturated with product from a previous similar run. The dried product, amounting to 987 grams, was a white crystalline solid having a melting range of 143 degrees to 147 degrees centigrade. The product was recrystallized from petroleum ether to obtain the product having a melting point of 146-147 degrees centigrade.

One hundred grams of the product so-prepared was then added to a solution of potassium hydroxide (300 grams) in methanol (850 milliliters) at a substantially uniform rate over a period of 35 minutes. The temperature of the vigorous reaction was maintained at 55-65 degrees centigrade and the reaction product was diluted to two liters with water to form a clear solution. The diluted reaction product was then acidified with hydrochloric acid and a bulky, gelatinuous, white precipitate obtained thereby. The solid was filtered, dissolved in ethyl ether, some water was separated, and discarded, and the ether layer was evaporated to obtain a white solid. Carbon tetrachloride was added to the solid and the mixture evaporated to remove the remaining water. The crystals, after drying and recrystallization from carbon tetrachloride-acetone, sublimed at high temperature and had a sealed tube melting point of about 355 degrees centigrade.

Photostat copies are attached of pages numbered 1, 4, 5, 6, and 8 from Progress Report No. 12 of Jack S. Newcomer and E. T. McBee, prepared by them in the ordinary course of their researches in the laboratories of the Chemistry Department of Purdue University at Lafayette, Indiana, evidencing the foregoing.

Further, affiants sayeth not.

/s/ Earl T. McBee

Plaintiff's
Exhibit 1

State of Indiana)
: ss.
County of Tippecanoe)

Before me personally appeared Earl T. McBee, to me personally known, and being duly sworn, made oath before me to the allegations set forth herein on this 12th day of April, 1954.

/s/ Phyllis Anne Hebein
Notary Public

[SEAL]
My Commission Expires: 2/6/57

/s/ Jack S. Newcomer

State of New York)
: ss.
County of Niagara)

Before me personally appeared Jack S. Newcomer, to me personally known, and being duly sworn, made oath before me to the allegations set forth herein on this 13 day of April, 1954.

/s/ Alice Young
Notary Public

[SEAL]
My Commission Expires: March 30, 1956

Progress Report No. 12
December 25, 1957 to February 28, 1947

Hooker Electrochemical Company Fellowship No. 173

Purdue Research Foundation and Department of Chemistry
in cooperation with
Hooker Electrochemical Company
Jack S. Newcomer, Research Fellow
E. T. McBee, In direct charge

Discussion

YIELD STUDY OF J.N.-63

The encouraging results of preliminary insecticidal tests on J.N.-63, obtained by the action of chlorosulfonic acid on hexachlorocyclopentadiene, have prompted a yield study of this reaction. The conversion has been increased progressively from 44.3 to 45.3, 66.8, and 90.9%. The yield in the latter experiment was 95.5%. A brief summary of these experiments is presented in Table I.

Plaintiff's
Exhibit 1

Table I

Action of $ClSO_3H$ on C_5Cl_6

Exp. No.	Conv.,%	Yield,%	Conditions Temp., C.	 Time, hrs.
J.N.-63	44.3	---	65-75	1.75
161	45.3	90.9	75	2.75
162	66.8	93.7	75	5.0
163	90.9	95.5	90	2.0

The yield data are based upon the quantity of hexachlorocyclopentadiene used and recovered. In each experiment, the product was filtered from the reaction slurry, thus producing a filtrate containing chlorosulfonic acid, unreacted hexachlorocyclopentadiene, and some J.N.-63. As demonstrated by Exp. No. J.N.-163, the filtrate may be processed by either of two methods. Method A involved the decomposition of the filtrate by

* * *

Exp. No. J.N.-161. Preparation of $(C_{10}Cl_{12}SO_3)ClH$

The first preparation of $(C_{10}Cl_{12}AO_3)_xClH$ (Exp. No. J.N.-63) was performed by vigorously stirring a mixture of 500 g. (1.84 moles) of C_5Cl_6 and 1,000 g. (8.62 moles) of $ClSO_3H$ for 1.75 hrs. at 65-75°. The conversion was 44.3%. Exp. No. J.N.-161 was performed by vigorously stirring a mixture of 1,000 g. (3.68 moles) of C_5Cl_6 and 1,000 g. (8.62) moles of $ClSO_3H$ at 75° for 2.5 hrs. At this time, red crystals had just begun to crystallize from the reaction mixture, which is a clear, maroon color. Oxides of sulfur are liberated throughout the reaction. Upon cooling the reaction mixture to 3°, sufficient red crystals had separated to form a thick slurry. Filtration and subsequent washing with water produced the white, crystalline product, which was contaminated with an appreciable quantity of C_5Cl_6. The C_5Cl_6 was removed by washing with 400 ml. pet. ether (90-100°). The dried product weighed 530 g. The $ClSO_3H$ filtrate consisted of two phases of about equal density. Partial separation gave a yellow liquid, which was combined with the yellow organic layer obtained from the aqueous filtrate.

Distillation produced 183 g. of C_5Cl_6. The remainder of the $ClSO_3H$ filtrate was decomposed with ice to give 204 g. of C_5Cl_6. The pet. ether filtrate was evaporated to give 103 g. of C_5Cl_6 with dissolved $(C_{10}Cl_{12}SO_3)ClH$. The conversion and yield were 45.3 and 90.9%, respectively.

Exp. No. J.N.-162. Preparation of $(C_{10}Cl_{12}SO_3)_xClH$

A mixture of 1,000 g. (8.62 moles) of $ClSO_3H$ and 1,000 g. (3.68 moles) of C_5Cl_6 was stirred vigorously for 5 hrs. at 75°C. Red crystals began to separate from the reaction mixture after 2.5 hrs. The product and excess reactants were recovered as in Exp. No. J.N.-161 to obtain 756 g. of $(C_{10}Cl_{12}SO_3)_xClH$, and 297 g. of C_5Cl_6. The conversion and yield were 66.8 and 93.7%, respectively.

Exp. No. J.N.-163. Preparation of $(C_{10}Cl_{12}SO_3)_xClH$

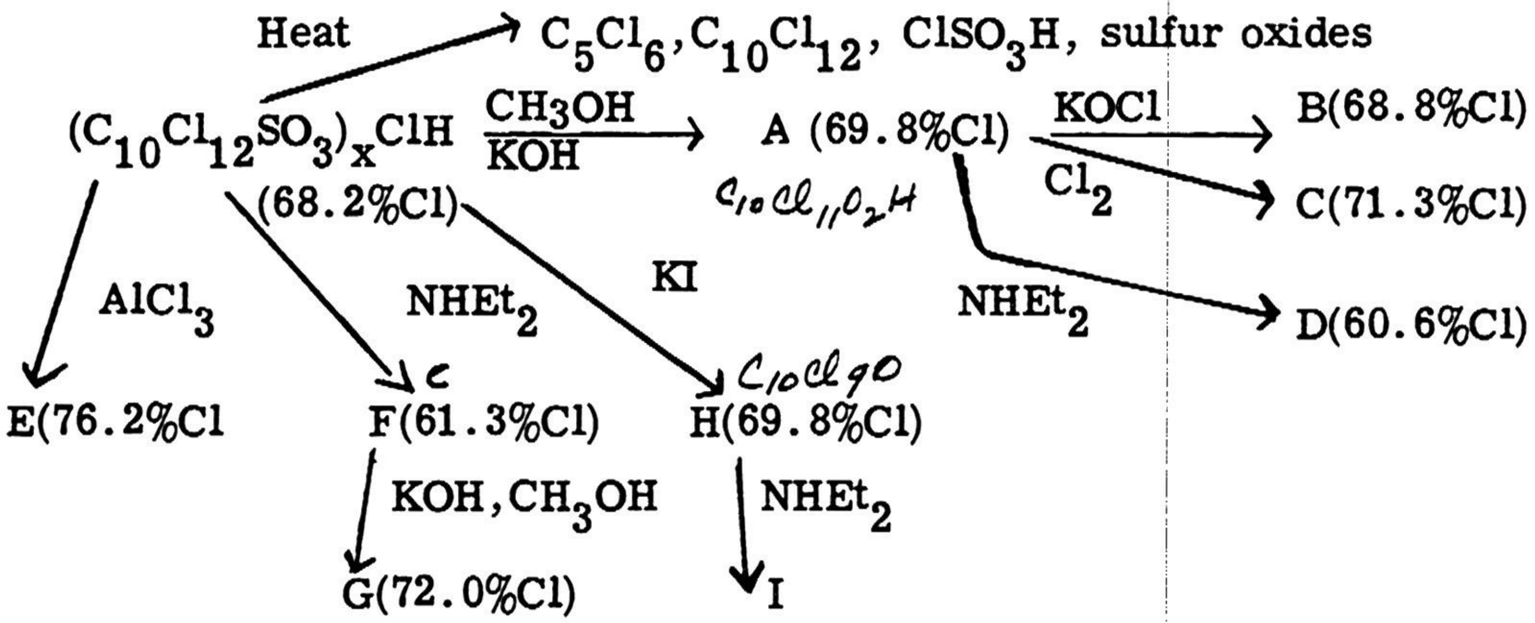
A mixture of 1,000 g. (8.62 moles) of $ClSO_3H$ and 1,000 g. (3.68 moles) of C_5Cl_6 was stirred vigorously for 2 hrs. at 90°C. Red crystals separated from the reaction mixture after 0.5 hr. The thick reaction mixture was cooled to 3° and maintained at that temperature for 2 hrs. The slurry was filtered through a sintered glass funnel to obtain the solid product, which was washed with water, and then with 400 ml. of pet ether (90-100°, saturated with $(C_{10}Cl_{12}SO_3)_xClH$. The dried product was a white, crystalline solid; 987 g., m.p. 143-147°. The $ClSO_3H$ filtrate consisted of one phase. No organic layer was present in the aqueous filtrate. Of the 804 g. of $ClSO_3H$ filtrate, 483 g. was distilled in a Claisen flask at 750 mm. to obtain 405 g. of clear, colorless $ClSO_3H$ (b.r. 138-152°). Thus, of the entire filtrate, 674 g. of $ClSO_3H$ is recoverable by this method. The rather large boiling range for the $ClSO_3H$ is attributed to the oxides of sulfur liberated during the initial portion of the distillation. These gases probably result from the action of $ClSO_3H$ on the organic material to form a tar, which is left as a residue. The tar does not react with water nor dissolve in it. It is significant that little or no sulfuric acid is formed in the reaction. These results show that 0.77 mole of $ClSO_3H$ is consumed per 1.00 mole of C_5Cl_6. Of the 0.77 mole of $ClSO_3H$, 0.50 is accounted for by the sulfur

Plaintiff's
Exhibit 1

in the product and 0.27 is not accounted for on a quantitative basis. However, the only loss of sulfur containing compounds that could have occurred would be in the gaseous by-products. It was noted that considerable evolution of oxides of sulfur occurred during the course of the reaction. Hence, one can predict, with reasonable degree of assurance, that approximately 0.27 mole of $ClSO_3H$ is accounted for as gaseous by-product. It may be significant that for each atom of sulfur in the product, 0.5 atom of sulfur is liberated as an oxide.

IDENTIFICATION STUDY OF J.N.-63-$(C_{10}Cl_{12}SO_3)_xClH$.

Chlorine and sulfur analyses on J.N.-63 indicated the formula $(C_{10}Cl_{12}SO_3)_x$. However, the isolation of chlorosulfonic acid from the pyrolytic products establishes the presence of hydrogen. On this basis, the formula is tentatively revised to $(C_{10}Cl_{12}SO_3)_xClH$. The following reactions have been performed.



Of considerable significance is the fact that none of the organic products contains sulfur. Products D, F, and I contain nitrogen. At present, the results obtained cannot be interpreted fully, although the increase in chlorine content and loss of sulfur in the preparation of A, E, and H, are of much significance considering the relatively high conversions that were obtained. These data show that $(C_{10}Cl_{12}SO_3)_xClH$ does not contain a sulfur linkage between C_5 units, but rather it contains a sulfur linkage with a C_{10} unit or between C_{10} units. Thus, the pyrolysis of J.N.-63 to hexachlorocyclopentadiene must occur by carbon-carbon fission. This is interesting in view of the stability of the dimer of hexachlorocyclopentadiene toward pyrolysis. The ease of elimination of sulfur suggests

* * *

Experiment No. J.N.-171. Action of alcoholic alkali on $(C_{10}Cl_{12}SO_3)_x$. Product A.

During a period of 35 min, 100 g. of $(C_{10}Cl_{12}SO_3)_xClH$ was added to a solution of 300 g. potassium hydroxide in 850 ml. methanol. The temperature of the vigorous reaction was maintained at 55-65°C. by the regulated addition of $(C_{10}Cl_{12}SO_3)_xClH$. The resulting solution was diluted to 2 1. with water, but no solid precipitated. Upon acidification with hydrochloric acid, an extremely bulky, gelatinous, white precipitate formed. After suction filtration and pressing, the resulting cake occupied the volume equivalent of a 4-1 beaker and about half that of a 3-1. beaker. The enormous amount of water was removed by dissolving the cake in ether and separating the ether from the aqueous layer. Evaporation of the ether layer gave a white solid which formed a gel upon initial attempts to crystallize it from carbon tetrachloride. The small amount of remaining water was removed by boiling a solution of the solid in carbon tetrachloride for about an hour. Recrystallization gave white crystals (64 g., sublime without melting at high temperature, 69.3,69.8%Cl). The product gave a negative qualitative test for sulfur.

Experiment No. J.N.-172 Action of diethylamine on $(C_{10}Cl_{12}SO_3)_xClH$. Product F.

Twenty grams of diethylamine was added dropwise to a solution of 15 g. $(C_{10}Cl_{12}SO_3)_xClH$ dissolved in 125 ml. of methanol. The resulting solution was diluted with water to precipitate the product, which was filtered and crystallized from carbon tetrachloride. The white needles (9g., 61.3, 61.5%Cl) decompose gradually above 150°C. without melting. The product gave a positive qualitative test for nitrogen, but a negative test for sulfur. This material tends to form a gel with water, but not

Plaintiff's
Exhibit 1

IN THE UNITED STATES PATENT OFFICE

Division : 38
Applicant : Earl T McBee et al
Serial : 238 186
Filed : 1951 July 23
For : COMPOSITION OF MATTER

Hon Commissioner of Patents
Washington 25 D C

AMENDMENT UNDER RULE 116

Sir:

In response to the Office Action mailed 1953 October 27, please amend as follows:

In the Specification:

Page 2, line 26: cancel "any"
Page 7, line 3: change "bases" to ---basis---.
Page 9, line 3: cancel "Upon cooling the pot residue to room".
line 4: cancel "temperature, the product solidified as a tan solid.".
Page 14, line 25: cancel "$C_{10}HCl_{10}O_2$" and insert therefor ---$C_{10}H_2Cl_{10}O_2$---.
Page 14, line 30: Cancel "$C_{10}HCl_{10}O_2$" and insert therefor ---$C_{10}H_2Cl_{10}O_2$---.
Page 15, line 2: cancel "$C_{10}Cl_{10}O_2K$" and insert therefor ---$C_{10}Cl_{10}O_2KH$---.
line 3: cancel "$C_{10}HCl_{10}O_2$" and insert therefor ---$C_{10}H_2Cl_{10}O_2$--.
line 11: cancel "$C_{10}HCl_{10}O_2$" and insert therefor ---$C_{10}H_2Cl_{10}O_2$---.
line 13: cancel "$C_{10}HCl_{10}O_2$" and insert therefor ---$C_{10}H_2Cl_{10}O_2$---.
line 14: cancel "$C_{10}HCl_{10}O_2$" and insert therefor ---$C_{10}H_2Cl_{10}O_2$---.

Page 15, line 20: cancel "$C_{10}HCl_{10}O_2$" and insert therefor ---$C_{10}H_2Cl_{10}O_2$---.

line 22: cancel "$C_{10}HCl_{10}O_2$" and insert therefor ---$C_{10}H_2Cl_{10}O_2$---.

line 28: cancel "$C_{10}HCl_{10}O_2$" and insert therefor ---$C_{10}H_2Cl_{10}O_2$---.

line 29: cancel "$C_{14}H_{12}Cl_{10}O_2N$" and insert therefor ---$C_{14}H_{13}Cl_{10}O_2N$---.

Page 16, line 1: cancel "$C_{10}HCl_{10}O_2$" and insert therefor ---$C_{10}H_2Cl_{10}O_2$---.

line 8: cancel "$C_{10}HCl_{10}O_2$" and insert therefor ---$C_{10}H_2Cl_{10}O_2$---.

line 11: cancel "$C_{10}HCl_{10}O_2$" and insert therefor ---$C_{10}H_2Cl_{10}O_2$---.

line 12: cancel "$C_{10}HCl_{10}O_2$" and insert therefor ---$C_{10}H_2Cl_{10}O_2$---.

line 13: cancel "$C_{10}HCl_{10}O_2$" and insert therefor ---$C_{10}H_2Cl_{10}O_2$---.

line 21: cancel "$C_{10}HCl_{10}O_2$" and insert therefor ---$C_{10}H_2Cl_{10}O_2$---.

line 27: cancel "$C_{10}HCl_{10}O_2$" and insert therefor ---$C_{10}H_2Cl_{10}O_2$---.

line 28: cancel "$C_{10}HCl_{10}O_2$" and insert therefor ---$C_{10}H_2Cl_{10}O_2$---.

line 29: cancel "$C_{10}HCl_{10}O_2$" and insert therefor ---$C_{10}H_2Cl_{10}O_2$---.

In the Claims:

Claims 5-8, inclusive, are all of the claims presently pending in the above-identified application.

Please insert the following claim for purposes of appeal:

Plaintiff's
Exhibit 1

A composition of matter which has the following characteristics: it is a white crystalline solid having a sealed tube corrected melting point of about 355 degrees centigrade, it behaves as a weak organic acid, is very soluble in polar organic solvents, substantially insoluble in non-polar organic solvents and has the approximate formula $C_{10}Cl_{10}O$ and is preparable from a compound having a melting point of approximately 146-147 degrees centigrade, formed by the reaction of hexachlorocyclopentadiene with chlorosulfonic acid, by treatment with alcoholic alkali and acidification and dehydration.

REMARKS

The interview of 1954 April 16 is acknowledged with thanks. At the interview it was agreed the above changes in the specification would be acceptable. It was also agreed proposed Claim 13 would be entered for purposes of appeal.

The proposed claim follows substantially the language of the claim indicated as probably allowable in the Advisory Office Action mailed 1953 September 30 and is supported, as to characterization of the composition, particularly at page 1, line 20, et seq., of the specification, and as to the method whereby it can be prepared by preparation 1 as described on page 11, beginning at line 11, and in Example 5, beginning at line 5 of page 12. Reference is made for further description of the starting compound to Patent 2 516 404 of the same applicants and page 11, line 29, et seq., of the specification.

A Rule 131 affidavit is submitted herewith antedating 1950 November 7, the filing date of Gilbert et al 2 616 928, the only reference now standing in the application.

It is clear from inspection of Applicants' patent 2 516 404 that Applicants had reacted alcoholic alkali with the compound of patent Claim 7, and acidified the same and dehydrated it during sublimation, see column 3, line 45 et seq. It would thus appear that Applicants clearly reduced to practice the subject matter of proposed Claim 13 and this disclosure in their prior patent is a positive indication of invention by Applicants before the filing date of the Gilbert et al reference.

Claim 13 is submitted to be definite. The product is characterized by properties and empirical formula, and a procedure is set forth for making it. The fact that it may be prepared by other procedures, as evidenced by description of several such procedures in the specification of Applicants, it is not considered material on the issue of accurately defining the product whereby the public may know the precise limits of the subject matter from which it would temporarily be precluded by the grant of the claim to Applicants. The situation here is submitted to be controlled by Ex parte Smith, 43 USPQ 157 and not by Ex parte Lighty, 64 USPQ 430.

Entry of Claim 13 is solicited for purposes of appeal.

The claims in the application are 5, 6, 7, 8 and 13.

Respectfully

/s/ Dean Laurence
Attorney

1954 April 19

[Mailed May 5, 1954, Pat. Div. 38]

DEPARTMENT OF COMMERCE
UNITED STATES PATENT OFFICE
Washington

Paper No. 14

Dean Laurence
Box 151
One Clinton Ave.
St. Johns, Mich.

Applicant: Earl T. McBee et al
Ser. No. 238,186
Filed July 23, 1951
For COMPOSITION OF MATTER

Please find below a communication from the EXAMINER in charge of this application.

Robert C. Watson
Commissioner of Patents.

Plaintiff's
Exhibit 1

Responsive to amendment and affidavit filed Apr. 21, 1954.

Applicants' amendment under Rule 116 (Apr. 21, 1954) has been entered since it is considered to place the instant case in better condition for appeal. The Examiner notes that Applicants have appealed under Rule 191 from the final rejection of claims 5, 6, 7, 8 and that appeal is taken on newly-submitted claim 13. Because of a typographical error committed in the final rejection of Oct. 27, 1953, Applicants were given to believe that the amendment dealt therewith had not been entered, that claims 5-8 remained in the case and that the amendments to the specification had not been entered. This conclusion is further substantiated by the fact that Applicants have reiterated said amendments to the specification in the above-mentioned amendment under Rule 116 (Apr. 21, 1954). In view of this confusion and in order to clarify the record, it is felt it would be best to treat the amendment dated Oct. 12, 1953, paper No. 8/D as not having been entered.

Such a procedure would relegate the record to the final rejection of June 17, 1953, in which claims 5-8 were rejected as indefinite and improper product claims. Newly-added claim 13, mentioned above, is also subject to the same grounds of rejection. It is the Examiner's opinion that the present action clarifies an otherwise inconsistent record.

Applicants' affidavit under Rule 131 is considered adequate to antedate the Gilbert et al reference (2,616,928) relied on in the Office action dated Oct. 27, 1953. Accordingly, said reference is no longer relied on as anticipatory of the present claims.

FWGuay/aw Examiner

Plaintiff's
Exhibit 1

[Mailed July 27, 1954, Pat. Div. 38]

DEPARTMENT OF COMMERCE
UNITED STATES PATENT OFFICE
Washington

Paper No. 16

In re application of	)	
Earl T. McBee et al.	)	Appeal No. 423,72
Ser. No. 238,186	)	Before the Board of Appeals
Filed July 23, 1951	)	
For COMPOSITION OF MATTER	)	

Mr. Dean Laurence for appellants.

EXAMINER'S ANSWER

This is an appeal from the final rejection of claims 5-8 and 13.

A corrected copy of the appealed claims appears on page 1 of applicants' brief.

The final rejection is not based on prior art but on indefiniteness and impropriety.

DESCRIPTION OF THE INVENTION

Hexachlorocyclopentadiene has been reacted with sulfur trioxide to form a compound of the approximate molecular formula $C_{10}H_2O_3SCl_{12}$ having a melting point, in the solid state, of about 146 to 147 degrees centigrade (cf. claim 7, U.S. Pat. 2,516,404 granted to present applicants). Applicants have now discovered that said compound having a m.p. of about 146-7 degrees centigrade may be used to prepare new and useful compounds. These new compounds are now being claimed in this application which also discloses several methods for preparing same.

Plaintiff's
Exhibit 1

BASIS FOR FINAL REJECTION

Applicants have attempted to claim their compounds by reciting their characteristics (see claims 5-8) or by reciting both the characteristics and a process which may be employed to obtain products having said characteristics (see claim 13). Claims 5-8 have been rejected as being indefinite and improper product claims. Since the structure of the product is not known, it must be defined by its method of manufacture. Ex parte Fesenmeier, 1922 C.D. 18; Ex parte Serini et al., 68 U.S.P.Q. 282. Claim 13 is subject to the same rejection and is further objectionable in the language "is preparable from," which language is considered indefinite and improper in that it fails to limit the methods employable to produce the desired product. Consequently, claim 13 is considered as indefinite as claims 5-8.

ARGUMENTS

Claims 5-8 and 13 are deemed indefinite and improper for the reason that the product characteristics as recited are not considered adequate to particularly define said product. Expressions such as " it behaves as a weak organic acid," "is substantially insoluble in non-polar organic solvents," etc. are not regarded as definitive enough to aid in supplying a composite picture of the compound claimed. Applicants' arguments in their brief, pages 4 and 5, on this score have been carefully considered but are not deemed persuasive for the reason just given.

Claim 13 is rejected on the same grounds and for the same reason. Furthermore, the statement "is preparable from" renders said claim unlimited in scope as to the processes of preparing the compounds in question. As such, claim 13 is not deemed a true product by process claim; in fact, it differs from claims 5-8 only in suggesting a method for preparing the compounds. Arguments by applicants to the effect that their specification discloses several processes for preparing the instant compounds do not appear to provide proper support for a claim that reads on any method of preparing the instant compounds. It would seem to be extending the doctrine of Ex parte Smith (cf. pages 8-11, applicants' brief) to draw an analogy between the expressions "substantially identical with" and "is preparable from" since the former phrase does not render the methods of preparing the product unlimited, i.e., there is but one method recited for obtaining the product. Another objection to claim 13 is that while the phrase "is preparable from" resembles a modification of the scope it actually does not modify it whatsoever. The instant case is also readily distinguishable from Ex parte Smith in that the claims in the latter teach the characteristics of the product claimed as well as a definite method for preparing same. Claim 13 is not so limited as to the method for preparing the product. Ex parte Lichty, 64 U.S.P.Q. 430, particularly refers to the fact that the phrase "substantially identical with" in the claims of Ex parte Smith is not necessary in said claims, but that said phrase is additional matter. In short, Ex parte Lichty recognized a distinction with a significance, the significance being that the phrase in question did not render the claim indefinite. In the last analysis, it is the Examiner's position that "is preparable from" renders claim 13 indefinite and improper.

It is respectfully submitted that the above rejection is sound and should be affirmed.

/s/ F. W. Guay
Examiner, Div. 38

* * *

DEPARTMENT OF COMMERCE
UNITED STATES PATENT OFFICE
Washington

Paper No. 17
December 16, 1954

Dean Laurence
Box 151
One Clinton Ave.
St. Johns, Michigan

Plaintiff's
Exhibit 1

NOTICE OF HEARING

The case of Earl T. McBee et al.

Appeal No: 423-72

Serial No.: 238,186

will be heard by the Board of Appeals on the

8th day of December, 1955.

The hearings will commence at 9:30 A.M. and as soon as the argument in one case is concluded, the succeeding case will be taken up.

The time allowed for argument is thirty minutes.

By special leave, obtained before the argument is commenced, the time may be extended.

Very truly yours,

/s/ Robert C. Watson
Commissioner of Patents

PLEASE REFER TO APPEAL NUMBER IN ALL COMMUNICATIONS CONCERNING THIS CASE

IN THE UNITED STATES PATENT OFFICE

Division : 31
Applicant : Earl T. McBee et al.
Serial : 238 186
Filed : 1951 July 23
For : COMPOSITION OF MATTER

1955 December 6
FDL:160:DL:kw
PRF 4-1-517

Honorable Commissioner of Patents
Washington 25, D. C.

Sir:

ASSOCIATE POWER OF ATTORNEY UNDER RULE 34

Dean Laurence, being the attorney of record in the above-identified application, hereby appoints Winston E. Miller (Registration Number 17 054) whose address is Saint Johns, Michigan, as associate, to inspect the file, to prepare and file amendments, and to conduct all business in the Patent Office relating to the prosecution of the said application.

PLEASE ADDRESS ALL COMMUNICATIONS TO DEAN LAURENCE, SAINT JOHNS, MICHIGAN.

Respectfully,

/s/ Dean Laurence
Attorney

[Mailed December 19, 1955, U. S. Patent Office, Board of Appeals]

Paper No. 19

Appeal No. 423-72

Hearing:
December 8, 1955

IN THE UNITED STATES PATENT OFFICE

BEFORE THE BOARD OF APPEALS

Ex parte Earl T. McBee and
Jack S. Newcomer

Application for Patent filed July 23, 1951, Serial No. 238,186.
Composition of Matter.

Dean Laurence and Winston E. Miller for appellants.

This is an appeal from the action of the examiner finally rejecting claims 5, 6, 7, 8 and 13, which are all of the claims pending in the case.

Claim 13 is reproduced as illustrative.

13. A composition of matter which has the following characteristics: it is a white crystalline solid having a sealed tube corrected melting point of about 355 degrees centigrade, it behaves as a weak organic acid, is very soluble in polar organic solvents, substantially insoluble in non-polar organic solvents and has the approximate formula $C_{10}Cl_{10}O$ and is preparable from a compound having a melting point of approximately 146-147 degrees centigrade, formed by the reaction of hexachlorocyclopentadiene with chlorosulfonic acid, by treatment with alcoholic alkali and acidification and dehydration.

Plaintiff's
Exhibit 1

The appealed claims are addressed to an assertedly novel compound and the sole question presented is that of adequacy of definition of that compound in accordance with 35 U.S. Code (1952 Ed.) Section 112.

Appellants have discussed appealed claims 6 and 13 as typical and as determinative of the issues here presented. Claim 6 is identical with the first portion of claim 13 as written above, down to and including the formula in line 7. Appellants refer to claim 6 as defining the melting point and the molecular or empirical formula of the compound. It is asserted that organic compounds may be sufficiently identified by such properties. The claim also refers in very general language to the solubility of the compound and includes the statement that the compound is similar in some properties to organic acids. The record of this case does not warrant an assumption that the melting point and molecular formula are significant identifying elements. The claim refers to the melting point as "about 355^{O}C." The table on page 6 of the specification gives figures obtained in determining the melting point of several samples of the claimed compound. These figures range from 331 to 351 for the first softening point; 351 to 355 for the meniscus, and 352 to 361 for the first crystals. It appears therefore that the qualifying term "about" is intended to include a variation of up to 5^{O} above or below the figures specified. We do not believe that it can be seriously asserted that a melting point range as wide as indicated defines an identifying characteristic.

The molecular formula in the claim refers to the presence of 10 carbon atoms, 10 chlorine atoms and 1 oxygen atom. The original specification stated the formula as also including an additional atom of hydrogen and of oxygen. Since the molecular formula now specified in claim 6 is not derivable from the original specification, it cannot be asserted that the claimed formula constitutes essential identifying information. The statement of the molecular formula is qualified by the term "approximate". Qualifying terms of this character are normally employed in claims to compositions of matter and are generally acceptable as providing for insignificant variation from particularly named limiting amounts. The reasoning ordinarily employed in justifying such qualifying terms is inapplicable to a molecular formula since a molecular formula represents a stated number of atoms of each of the kinds indicated and there can be no variation by a fractional portion of an atom. The word "approximate" used in connection with the formula specified in claim 6 can only have significance as indicating that the formula may be varied by a whole number of any one or more of the three kinds of atoms indicated. Such a variation wholly destroys any identifying significance in the formula. Since the molecular formula claimed was not originally disclosed, and since the statement of the molecular formula in the claim is qualified in a manner to render such formula meaningless, we must hold that statement in the claim in this respect is without identifying significance.

The general statement of solubility, as very soluble in polar organic solvents,and substantially insoluble in non-polar organic solvents, is so broad as to obviously include many compounds of entirely different characteristics. The same observation is applicable to the expression "behaves as a weak organic acid". There is no assertion the compound is in fact an organic acid and the statement in question is indefinite. It is accordingly our conclusion that claim 6 is properly rejected as not adequately defining the subject matter involved.

Claim 13 differs from claim 6 in the addition of a statement in general language of the procedure required to prepare the claimed compound and introduced with the term "is preparable from". The examiner holds that this statement of process is not an adequate statement of the process and moreover does not confine the claim to products produced by the process. The general rule suggested in Ex parte Fesenmeier 1922 C.D. 18; 302 O.G. 199 to the effect that a product may be defined in terms of the process of preparing it ordinarily requires the claim to be couched in such terms as to adequately define the process and confine the claimed subject matter to products of the process. This is a generous rule which permits an inventor to claim a product which he is unable to adequately identify apart from the process. To permit claims in broad enough language to include products of similar character prepared by other processes would in general require workers in the art to determine the identity of the product where the inventor himself has been unable or unwilling to do so. Appellants rely on Ex parte Smith, 43 U.S.P.Q. 157 in support of their position with respect to the use of the term "preparable from". The examiner relies on Ex parte Lichty, 64 U.S.P.Q. 430 and Ex parte Serini et al., 68 U.S.P.Q. 282 in support of a contrary position.

We have studied these decisions with care. The Ex parte Smith case represents an exceptional situation in which it appears that the balance of the claim substantially identified and defined the novel composition, the added statements concerning process being unnecessary from the standpoint of such identification. We are of the opinion that the decisions cited by the examiner provide a more adequate statement of the law applicable to situations of this character. Claim 13 is therefore in substance a combination of two different and inadequate definitions of the novel chemical compound. We have sufficiently discussed the statements of the characteristics in connection with our discussion of claim 6. The process recited in claim 13 is not sufficiently definite to insure the preparation of the chemical compound in question and the further broadening of the claim 6 referring to compounds of a character which are "preparable" by the stated process would place the burden of adequately identifying the compound, which appellants have been unwilling or unable to identify, on other workers in this art. The rejection of claim 13 as indefinite and not adequately defining the subject matter will be sustained.

It is apparent from the terms of the claims themselves that claims 5, 7 and 8 are subject to the same considerations as claim 6.

The decision of the examiner is affirmed.

/s/ E. W. Geniesse)
Examiner-in-Chief)
)
/s/ C. S. Duncombe) BOARD
Examiner-in-Chief) OF
) APPEALS
/s/ B. Henkin)
Examiner-in-Chief)
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December 19, 1955

Patented July 25, 1950 **2,516,404**

UNITED STATES PATENT OFFICE

2,516,404

REACTION PRODUCT OF HEXACHLORO-CYCLOPENTADIENE AND CHLORSULFONIC ACID

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Serial No. 1,486

7 Claims. (Cl. 260—1)

The present invention relates to a novel composition of matter, the compound of the molecular formula $C_{10}H_2O_3SCl_{12}$, and to a method for the preparation thereof by the reaction of chlorosulfonic acid and hexachlorocyclopentadiene. The new compound, having the approximate molecular formula $C_{10}H_2O_3SCl_{12}$, is a powerful insecticide, showing exceptional activity against the milkweed bug and the sweet potato weevil as well as other undesirable insects.

It is an object of the present invention to provide a novel composition of matter, the compound having the approximate formula $C_{10}H_2O_3SCl_{12}$. Another object of the present invention is the provision of a process for the preparation of the novel compound of the approximate formula $C_{10}H_2O_3SCl_{12}$. An additional object of the invention is a provision of a method for the preparation of the novel compound $C_{10}H_2O_3SCl_{12}$ from hexachlorocyclopentadiene. A still further object of the present invention is the provision of a process for the preparation of the novel insecticide $C_{10}H_2O_3SCl_{12}$ by the reaction of hexachlorocyclopentadiene and chlorosulfonic acid. Other objects of the invention will become apparent hereinafter.

The method of the present invention essentially comprises the reaction of hexachlorocyclopentadiene and chlorosulfonic acid. The reaction may be performed by mixing the reactants at room temperature and heating to the most desirable reaction temperature; by adding the chlorosulfonic acid to the hexachlorocyclopentadiene at a desirable reaction temperature; or by adding hexachlorocyclopentadiene to the chlorosulfonic acid at a desirable reaction temperature. Any suitable manner of admixing the reactants may be employed, and any variation of the above steps is likewise suitable procedure. Agitation of the reaction mixture is preferably employed to ensure satisfactory contact of the reactants, and is desirable for the attainment of highest yields of product.

The temperature range for the reaction between hexachlorocyclopentadiene and chlorosulfonic acid is anywhere between about 30 and 170 degrees centigrade, with the range between about 70 and 120 degrees centigrade being preferred. At temperatures below about 45 degrees centigrade, the reaction rate is quite slow, while at temperatures of about 75 degrees centigrade, a conversion of about 67 per cent may be obtained in a period of 5 hours. At a temperature of about 90 degrees centigrade, a 91 per cent conversion and a 96 per cent yield have been obtained in 2 hours. Although the product may also be obtained in good yield at temperatures of 120 to 130 degrees centigrade, the reaction rate at 85 to 95 degrees centigrade is rapid enough for all practical purposes. The time allowed for the reaction is relatively unimportant, the reaction appearing to be very rapid. Crystals of reaction product usually begin to separate from the mixture of reactants after about the first one-half hour. The reaction may be continued until substantially all of the starting compound, which is introduced into the reaction in the smaller quantity, is converted to the desired product.

Although only approximately 0.77 mole of chlorosulfonic acid appears to be consumed per each mole of hexachlorocyclopentadiene, it is usually advantageous to employ at least about 500 grams, preferably about 1000 grams, of chlorosulfonic acid for every 1000 grams of hexachlorocyclopentadiene, as, in this manner, fluidity of the reaction mixture is conveniently maintained. Thus, the mole ratio of chlorosulfonic acid to hexachlorocyclopentadiene may be varied from 0.77 to 1.0 to several moles of chlorosulfonic acid for each mole of hexachlorocyclopentadiene. However, an excess of the chlorosulfonic acid is usually employed. Other ratios of chlorosulfonic acid and hexachlorocyclopentadiene may be used if desired and, instead of introducing the chlorosulfonic acid directly into the reaction as such, it may be produced in situ.

An inert medium may also be employed for the reaction, although such is not necessary if an excess of chlorosulfonic acid is used. The reaction may be conducted conveniently without the presence of a solvent, and therefore, while a solvent may be employed, such is usually only used when it is desired not to employ an excess of chlorosulfonic acid.

The products of the reaction may be recovered by cooling the thick reaction product, filtering the slurry through a sintered glass funnel to obtain the solid product, washing with water, and then with petroleum ether or other suitable solvent which is saturated with the desired product. For purification purposes, the product may be conveniently recrystallized from petroleum ether, having a boiling range between about 30 and 100 degrees centigrade, or in other conventional manner.

The following examples are given to illustrate the practice of the present invention, but are in no way to be construed as limiting.

Example 1

A mixture of 1000 grams of chlorosulfonic acid and 1000 grams of hexachlorocyclopentadiene was stirred vigorously for 2 hours at 90 degrees centigrade. Red crystals began to separate from the reaction mixture after 0.5 hour. The thick reaction mixture was cooled to 3 degrees centigrade and maintained at that temperature for 2 hours. The slurry was filtered through a sintered glass funnel to obtain the solid product, which was washed with water, and then with 400 milliliters of petroleum ether saturated with $C_{10}H_2O_3SCl_{12}$. The dried product, 987 grams, was a white crystalline solid having a melting range of 143 to 147 degrees centigrade, and having the probable molecular formula $C_{10}H_2O_3SCl_{12}$. From the initial filtrate, 674 grams of chlorosulfonic acid was recovered by distillation. Recrystallization of the organic product from petroleum ether increased the melting point to 146 to 147 degrees centigrade.

Example 2

A mixture of 1000 grams of chlorosulfonic acid and 1000 grams of hexachlorocyclopentadiene was stirred for 5 hours at 75 degrees centigrade. Recovery of the product, performed as described previously, produced 756 grams of white solid, having the probable molecular formula $C_{10}H_2O_3SCl_{12}$.

Properties of $C_{10}H_2O_3SCl_{12}$

This material is an odorless, white, crystalline solid having a melting point of 146 to 147 degrees centigrade. It was found to contain 19.06 per cent carbon, 0.26 per cent hydrogen, 67.8 per cent chlorine, and 5.09 per cent sulfur. Molecular weight determinations gave the values 611 and 639. These data correspond closely to the molecular formula $C_{10}H_2O_3SCl_{12}$, which has the following analysis: C, 19.11; H, 0.32; Cl, 67.8; S, 5.09; M. W., 627. The product is soluble in common organic solvents, a saturated solution in petroleum ether (90–100 degrees centigrade) containing approximately 6 per cent of $C_{10}H_2O_3SCl_{12}$. It contains chlorine active toward silver nitrate and various alkaline reagents. For example, alcoholic alkali converts it to a water-soluble salt, which, upon acidification, forms a white crystalline acid having a very marked tendency to form a gel with water. This acid contains about 70 per cent chlorine, no sulfur, and has the property of subliming without melting at 740 millimeters of mercury pressure. The acid was converted to a compound containing nitrogen, and about 60 per cent chlorine by reaction with diethylamine. The nitrogen-containing product does not have a sharp melting point, but decomposes gradually at temperatures about about 160 degrees centigrade. The compound, $C_{10}H_2O_3SCl_{12}$, also undergoes reactions with alcohols, such as methyl and ethyl alcohol and also with the following reagents to produce products as described:

Reagent	Properties of Product
(a) Potassium iodide	White solid, sublimes without melting. Contains about 70 per cent chlorine, no sulfur.
(b) Diethylamine	White solid. Decomposes above 160 degrees centigrade. Contains nitrogen, about 61 per cent chlorine, no sulfur.
(c) Aluminum Chloride	White solid. Sublimes without melting. Contains 76 per cent chlorine, no sulfur.
(d) Application of heat to effect a distillation.	Hexachlorocyclopentadiene, chlorosulfonic acid, sulfur oxides, and the dimer of hexachlorocyclopentadiene.

The compound $C_{10}H_2O_3SCl_{12}$ is highly effective in 0.5 per cent concentrations against the milkweed bug and the sweet potato weevil, producing 100 per cent kills in 48 hours. It has also shown activity against German roaches and aphids. The material has been formulated as a wettable dust, as an emulsion, and in solutions of the solvents commonly used for formulating insecticides. At concentrations of 1:800, this insecticide has shown 0 per cent injury to greenhouse cucumbers, tomatoes, and beans.

The following is a tabulation of screening results on the new compound, conveniently referred to as JN–63:

Screening tests on JN–63

Comparison with High Gamma $C_6H_6Cl_6$	Actual Concentration	Milkweed Bug	Percent Mortality at 48 hours	
			Sweet Potato Weevil	Sweet Potato Weevil Sprayed
100..0..27	4:800, 0.5%	100	100	100
60..5..5	2:800, 0.25%	100	87	56
7..5..5	1:800, 0.13%	94	73	73
0..0..0	1:1600, 0.06%	56	53	56
	1:3200, 0.03%	46	15	0
	1:6400, 0.015%	20	0	0

Formulation	Actual Concentration	Insect used	Mortality at 48 hours
As a wettable dust	1:800	Milkweed Bug	100
Do	1:800	German Roaches	20
Do	1:800	Sweet Potato Weevil Sprayed.	73
Do	1:800	Sweet Potato Weevil placed on sprayed potato.	73
Do	1:1600	Milkweed Bug	90
As an emulsion	1:400	do	100
Do	1:800	do	100
Do	1:400	Sweet Potato Weevil	94
Do	1:800	do	93

JN–63 shows activity against aphids on celery plants at concentrations of 1:400 and 1:800.

Various modifications may be made in the method of the present invention without departing from the spirit or scope thereof, and it is to be understood that we limit ourselves only as defined in the appended claims.

We claim:

1. The process which includes: mixing together hexachlorocyclopentadiene and chlorosulfonic acid at a temperature above about room temperature, to cause reaction between the hexachlorocyclopentadiene and the chlorosulfonic acid.

2. The process of claim 1, wherein the reaction is conducted at a temperature between about 30 and about 170 degrees centigrade.

3. The process of claim 1, wherein the reaction is conducted at a temperature between about 70 and about 120 degrees centigrade.

4. The process of claim 1, wherein an excess of chlorosulfonic acid is employed.

5. The process of claim 1, wherein the product of the approximate composition $C_{10}H_2O_3SCl_{12}$ is separated from the reaction product.

6. A compound of the approximate molecular formula $C_{10}H_2O_3SCl_{12}$, having a melting point, in the solid state, of about 146 to 147 degrees centigrade, being substantially identical with the product formed by the reaction of chlorosulfonic acid with hexachlorocyclopentadiene and having valuable insecticidal properties.

7. A compound, having a melting point, in the solid state, of approximately 146 to 147 degrees centigrade, formed by the reaction of hexachlorocyclopentadiene and chlorosulfonic acid.

EARL T. McBEE.
JACK S. NEWCOMER.

REFERENCES CITED

The following references are of record in the file of this patent:

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Number	Name	Date
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2,148,928	Meuron	Feb. 28, 1939
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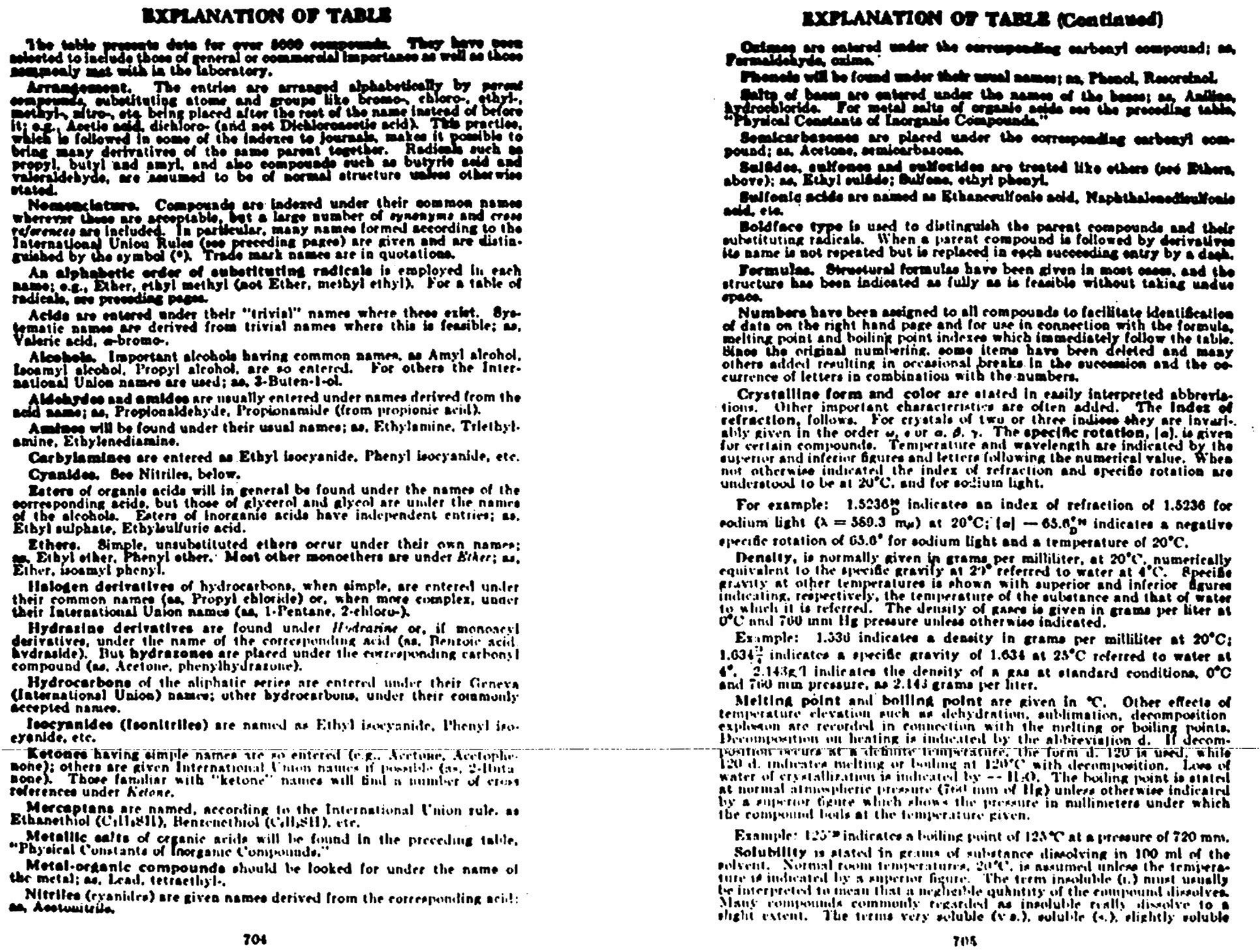
2,337,552	Henke	Dec. 28, 1943
2,382,038	Bruson	Aug. 14, 1945

FOREIGN PATENTS

Number	Country	Date
582,205	Great Britain	1946

PLAINTIFF's EXHIBIT 3

"The Handbook of Chemistry & Physics"
(Chem. Rubber Pub. Co.)



EXPLANATION OF TABLE

The table presents data for over 8000 compounds. They have been selected to include those of general or commercial importance as well as those commonly met with in the laboratory.

Arrangement. The entries are arranged alphabetically by parent compounds, substituting atoms and groups like bromo-, chloro-, ethyl-, methyl-, nitro-, etc. being placed after the rest of the name instead of before it; e.g., Acetic acid, dichloro- (and not Dichloroacetic acid). This practice, which is followed in some of the indexes to journals, makes it possible to bring many derivatives of the same parent together. Radicals such as propyl, butyl and amyl, and also compounds such as butyric acid and valeraldehyde, are assumed to be of normal structure unless otherwise stated.

Nomenclature. Compounds are indexed under their common names wherever these are acceptable, but a large number of *synonyms* and *cross references* are included. In particular, many names formed according to the International Union Rules (see preceding pages) are given and are distinguished by the symbol (°). Trade mark names are in quotations.

An alphabetic order of substituting radicals is employed in each name; e.g., Ether, ethyl methyl (not Ether, methyl ethyl). For a table of radicals, see preceding pages.

Acids are entered under their "trivial" names where these exist. Systematic names are derived from trivial names where this is feasible; as, Valeric acid, α-bromo-.

Alcohols. Important alcohols having common names, as Amyl alcohol, Isoamyl alcohol, Propyl alcohol, are so entered. For others the International Union names are used; as, 3-Buten-1-ol.

Aldehydes and amides are usually entered under names derived from the acid name; as, Propionaldehyde, Propionamide (from propionic acid).

Amines will be found under their usual names; as, Ethylamine, Triethylamine, Ethylenediamine.

Carbylamines are entered as Ethyl isocyanide, Phenyl isocyanide, etc.

Cyanides. See Nitriles, below.

Esters of organic acids will in general be found under the names of the corresponding acids, but those of glycerol and glycol are under the names of the alcohols. Esters of inorganic acids have independent entries; as, Ethyl sulphate, Ethylsulfuric acid.

Ethers. Simple, unsubstituted ethers occur under their own names; as, Ethyl ether, Phenyl ether. Most other monoethers are under *Ether*; as, Ether, isoamyl phenyl.

Halogen derivatives of hydrocarbons, when simple, are entered under their common names (as, Propyl chloride) or, when more complex, under their International Union names (as, 1-Pentane, 2-chloro-).

Hydrazine derivatives are found under *Hydrazine* or, if monoacyl derivatives, under the name of the corresponding acid (as, Benzoic acid hydrazide). But **hydrazones** are placed under the corresponding carbonyl compound (as, Acetone, phenylhydrazone).

Hydrocarbons of the aliphatic series are entered under their Geneva (International Union) names; other hydrocarbons, under their commonly accepted names.

Isocyanides (Isonitriles) are named as Ethyl isocyanide, Phenyl isocyanide, etc.

Ketones having simple names are so entered (e.g., Acetone, Acetophenone); others are given International Union names if possible (as, 2-Butanone). Those familiar with "ketone" names will find a number of cross references under *Ketone*.

Mercaptans are named, according to the International Union rule, as Ethanethiol (C_2H_5SH), Benzenethiol (C_6H_5SH), etc.

Metallic salts of organic acids will be found in the preceding table, "Physical Constants of Inorganic Compounds."

Metal-organic compounds should be looked for under the name of the metal; as, Lead, tetraethyl-.

Nitriles (cyanides) are given names derived from the corresponding acid; as, Acetonitrile.

EXPLANATION OF TABLE (Continued)

Oximes are entered under the corresponding carbonyl compound; as, Formaldehyde, oxime.

Phenols will be found under their usual names; as, Phenol, Resorcinol.

Salts of bases are entered under the names of the bases; as, Aniline, hydrochloride. For metal salts of organic acids see the preceding table, "Physical Constants of Inorganic Compounds."

Semicarbazones are placed under the corresponding carbonyl compound; as, Acetone, semicarbazone.

Sulfides, sulfones and sulfoxides are treated like ethers (see Ethers, above); as, Ethyl sulfide; Sulfone, ethyl phenyl.

Sulfonic acids are named as Ethanesulfonic acid, Naphthalenedisulfonic acid, etc.

Boldface type is used to distinguish the parent compounds and their substituting radicals. When a parent compound is followed by derivatives its name is not repeated but is replaced in each succeeding entry by a dash.

Formulas. Structural formulas have been given in most cases, and the structure has been indicated as fully as is feasible without taking undue space.

Numbers have been assigned to all compounds to facilitate identification of data on the right hand page and for use in connection with the formula, melting point and boiling point indexes which immediately follow the table. Since the original numbering, some items have been deleted and many others added resulting in occasional breaks in the succession and the occurrence of letters in combination with the numbers.

Crystalline form and **color** are stated in easily interpreted abbreviations. Other important characteristics are often added. The **index of refraction,** follows. For crystals of two or three indices they are invariably given in the order ω, ε or α, β, γ. The **specific rotation,** [α], is given for certain compounds. Temperature and wavelength are indicated by the superior and inferior figures and letters following the numerical value. When not otherwise indicated the index of refraction and specific rotation are understood to be at 20°C, and for sodium light.

For example: 1.5236$^{20}_{D}$ indicates an index of refraction of 1.5236 for sodium light (λ = 589.3 mμ) at 20°C; [α] — 65.6$^{\circ 20}_{D}$ indicates a negative specific rotation of 65.6° for sodium light and a temperature of 20°C.

Density, is normally given in grams per milliliter, at 20°C, numerically equivalent to the specific gravity at 20° referred to water at 4°C. Specific gravity at other temperatures is shown with superior and inferior figures indicating, respectively, the temperature of the substance and that of water to which it is referred. The density of gases is given in grams per liter at 0°C and 760 mm Hg pressure unless otherwise indicated.

Example: 1.330 indicates a density in grams per milliliter at 20°C; 1.634$^{25}_{4}$ indicates a specific gravity of 1.634 at 25°C referred to water at 4°. 2.143g/l indicates the density of a gas at standard conditions, 0°C and 760 mm pressure, as 2.143 grams per liter.

Melting point and **boiling point** are given in °C. Other effects of temperature elevation such as dehydration, sublimation, decomposition explosion are recorded in connection with the melting or boiling points. Decomposition on heating is indicated by the abbreviation d. If decomposition occurs at a definite temperature, the form d. 120 is used, while 120 d. indicates melting or boiling at 120°C with decomposition. Loss of water of crystallization is indicated by — H_2O. The boiling point is stated at normal atmospheric pressure (760 mm of Hg) unless otherwise indicated by a superior figure which shows the pressure in millimeters under which the compound boils at the temperature given.

Example: 125^{720} indicates a boiling point of 125°C at a pressure of 720 mm.

Solubility is stated in grams of substance dissolving in 100 ml of the solvent. Normal room temperatures, 20°C, is assumed unless the temperature is indicated by a superior figure. The term insoluble (i.) must usually be interpreted to mean that a negligible quantity of the compound dissolves. Many compounds commonly regarded as insoluble really dissolve to a slight extent. The terms very soluble (v s.), soluble (s.), slightly soluble

PLAINTIFF'S EXHIBIT 3

FORMULA INDEX OF ORGANIC COMPOUNDS
(Continued)

$C_9H_{14}O_6$, 2324, 4260.
$C_9H_{14}O_7$, 2794.
$C_9H_{15}NO$, 7420.
$C_9H_{15}NO_3\cdot H_2O$, 3412.
$C_9H_{15}NO_4$, 7033.
C_9H_{16}, 4450M, 6304, 6238T.
$C_9H_{16}ClNO_3$, 3413.
$C_9H_{16}O_2$, 197.
$C_9H_{16}O_4$, 951, 4214, 5424, 5445, 5447.
$C_9H_{17}ClO$, 6452.
$C_9H_{17}N$, 6451, 7664.
$C_9H_{17}NO\cdot H_2O$, 8549.
$C_9H_{17}NO_3$, 6415M.
C_9H_{18}, 3000, 3004, 6195.
$C_9H_{18}O$, 4493, 6190, 6191, 6192, B6560M.
$C_9H_{18}O_2$, 97, 2222, 2230, 2384, 3438, 4032, 4079, 4980, 4982, 5147, 6445, 8797, 8800.
$C_9H_{18}O_3$, 2567, 2571.
$C_9H_{19}N$, 7017M.
$C_9H_{19}NO$, 6445, 6446.
C_9H_{20}, 4461, 4463, 6178, 6255.
$C_9H_{20}N_2O$, 8765.
$C_9H_{20}O$, 3724, 4485, 4486, 4606, 6183, 6185, 6186, 6187, 6188, 6272, B3680J.
$C_9H_{20}O_2$, 6180.
$C_9H_{20}O_3$, 6328.
$C_9H_{20}O_6S_2$, 8165.
$C_9H_{21}B$, 1949.
$C_9H_{21}BO_3$, 7342.
$C_9H_{21}N$, 6198, 8660.
$C_9H_{21}O_4P$, B7367M.
$C_9H_{22}N_2S_2$, 2407.

C_{10}

$C_{10}H_4N_2O_4$, 5891, 5892, 5893.
$C_{10}H_5NO_{10}\cdot 3H_2O$, 7607.
$C_{10}H_5N_3O_6$, 5894, 5895, 5896, 5897.
$C_{10}H_6Br_2O$, 5988, 5991.
$C_{10}H_6ClNO_3$, 5845, 5846.
$C_{10}H_6Cl_2$, 5850, 5851, 5852, 5853, 5854, 5855, 5856, 5857, 5858, 5859.
$C_{10}H_6Cl_2O$, 5989.
$C_{10}H_6N_2O_3$, 7543.
$C_{10}H_6N_2O_4$, 5865, 5866, 5867.
$C_{10}H_6N_2O_5$, 5970, 5992
$C_{10}H_6O_2$, 6028, 6033, 6039.
$C_{10}H_6O_3$, 5172, 6031, 6032, 6037.
$C_{10}H_6O_3S$, 6015.
$C_{10}H_6O_4$, 4146, 5072, 5933.
$C_{10}H_6O_8$, 5504, 7084, 7557.
$C_{10}H_7Br$, 5840, 5841.
$C_{10}H_7BrO$, 5988.
$C_{10}H_7Cl$, 5842, 5843.
$C_{10}H_7ClO$, 5990.
$C_{10}H_7ClO_2S$, 5926, 5927.
$C_{10}H_7F$, 5870K, 5870M.
$C_{10}H_7I$, 5874, 5876.
$C_{10}H_7NO_2$, 2731M, 5883, 5884, 5973, 5974, 5997, 7620M, 7698K, 7698P, 7698R, 7698T, 7698V.
$C_{10}H_7NO_3$, 5279, 5971, 5972, 5994, 5995, 5996, 7416.
$C_{10}H_8$, 5833.
$C_{10}H_8Cl_4$, 5889.
$C_{10}H_8NNaO_3S$, 6017.
$C_{10}H_8N_2$, 1336, 1337, 1338, 1908.
$C_{10}H_8N_2O$, 6068.
$C_{10}H_8N_2O_2$, 6052, 6065, 6066, 6067.
$C_{10}H_8O$, 5955, 5978.
$C_{10}H_8O_2$, 2859, 2860, 5905, 5906, 5907, 5908, 5909, 5910, 5911, 5912, 5913, 5914.
$C_{10}H_8O_3$, 388.
$C_{10}H_8O_3S$, 5925.
$C_{10}H_8O_3S\cdot H_2O$, 5923.
$C_{10}H_8O_4$, 2755, 2756, 4138, 4149, 5433.
$C_{10}H_8O_4S$, 2907, 6008, 6009, 6010, 6011, 6013, 6014, 6016, 6018.
$C_{10}H_8O_6S_2$, 5916, 5919, 5920.
$C_{10}H_8O_7S_2$, 6005, 6006.
$C_{10}H_8O_9S_3$, 2702.
$C_{10}H_8S$, 5977, 6001.
$C_{10}H_9N$, 5332, 5043, 6059, 7622, 7679, 7681, 7682, 7683.
$C_{10}H_9NO$, 2593, 4882, 5960, 5961, 5962, 5963, 5964, 5982, 5983, 5984, 5985, 5986, 5987, 7077, 7702, 7705, 7707, 7709.
$C_{10}H_9NO_2$, 8019H, 84884M.
$C_{10}H_9NO_3S$, 6081, 6084, 6087, 6089, 6092.
$C_{10}H_9NO_3S\cdot 1\frac{1}{2}H_2O$, 5934.
$C_{10}H_9NO_4S\cdot H_2O$, 6083, 6085, 6086, 6088, 6090, 6091.
$C_{10}H_9NO_6$, 2772.
$C_{10}H_9NO_6S$, 6012.
$C_{10}H_9NO_7S_2$, 6073, 6074, 6075.
$C_{10}H_9NO_8S_2$, 6304.
$C_{10}H_{10}$, 1151, 5860.
$C_{10}H_{10}BrNO_3$, 191.
$C_{10}H_{10}ClN$, 6044, 6060.
$C_{10}H_{10}N_2$, 4725, 4726, 6095, 6096, 6097, 6098, 6099, 6100, 6101, 6102, 6138.
$C_{10}H_{10}N_2O$, 7457.
$C_{10}H_{10}N_2O_4$, B1540Q, B1540Y.
$C_{10}H_{10}N_2O_2S$, B8048T, B8050J.
$C_{10}H_{10}O$, 229.
$C_{10}H_{10}O_2$, 227, 385F, 1450, 2131, 2745, 2767, 5119, 7815.
$C_{10}H_{10}O_3$, 118, 2130, 7296, B4143J.
$C_{10}H_{10}O_4$, 2931, 3966, 4792, 5052, 5086, 5284, 5492, 6910, 6912, 7739, 8122, 8839.
$C_{10}H_{10}O_5$, 6314, 8108.
$C_{10}H_{10}O_6$, 4422, B5053M.
$C_{10}H_{11}N$, B4882M.
$C_{10}H_{11}NO_2$, 190, 3148, 3246.
$C_{10}H_{11}NO_3$, 318, 428.
$C_{10}H_{11}NO_4$, 8119R.
$C_{10}H_{12}$, 3211M, 6890.
$C_{10}H_{12}N_2$, 5073, 6128.
$C_{10}H_{12}N_2O$, 8718.
$C_{10}H_{12}N_2O_3$, 6537.
$C_{10}H_{12}N_2O_4$, 1015, 4271.
$C_{10}H_{12}O$, 563, 2102, 2103, 2270, 2923, 3479, 3656, 3657, 3658, 4997, 5975, 5976, 5999, 6000.
$C_{10}H_{12}O_2$, 1459, 1464, 1567, 1593, 1594, 1622, 2657, 2929, 3409, 3948, 5040, 5041, 5042, 5045, 7055, 8292, 8441, 8452, 8459, 8465.
$C_{10}H_{12}O_3$, 706, 1059, 2829, 4104, 7843, 8312T.
$C_{10}H_{12}O_4$, 2331, 8837.
$C_{10}H_{12}O_4S$, 8257.
$C_{10}H_{12}O_5$, 923, 1617, 1619.
$C_{10}H_{13}Br$, 3073.
$C_{10}H_{13}N$, 5173, 6056, 6070.
$C_{10}H_{13}NO$, 308, 310, 312, 314, 2218, 8167, 8285Q, 8285R.

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$C_{10}H_{14}BrN$, 596.
$C_{10}H_{14}NO_5$, 8290.
$C_{10}H_{14}N_2$, 558, 2206, 5074, 6129, 7008.
$C_{10}H_{14}N_2O$, 619.
$C_{10}H_{14}N_2O_3$, 617, 618, 6666, 6973.
$C_{10}H_{14}O$, 2605, 2616, 2930, 3682, 3753, 3775, 3776, 3777, 5038, 6711, 6712, 6713, 6714, 6716, 8288.
$C_{10}H_{14}O_2$, 1178, 1179, 1180, 6708, 6709, 6710, 7752, 7765, 8287.
$C_{10}H_{14}O_4$, 2322, 2323, 4147D, 4147L, 8799
$C_{10}H_{15}BrO$, 2311, 2312.
$C_{10}H_{15}ClO$, 2313.
$C_{10}H_{15}N$, 598, 599, 616, 647H, 647M, 685, 2607, 5039, 8205.
$C_{10}H_{15}NO$, 2615, 4672, 6739, 7411.
$C_{10}H_{15}NO\cdot H_2O$, 3451.
$C_{10}H_{15}NO_2$, 2314.
$C_{10}H_{15}N_3O$, 6975.
$C_{10}H_{16}$, 1962, 2301, 2302, 3964, 5355, 5356, 5810, 6219, 6219B, 6605, 6606, 6991, 7810, 8090, 8134, 8139.
$C_{10}H_{16}ClNO$, 3452, 7412.
$C_{10}H_{16}Cl_2N_2$, 6130.
$C_{10}H_{16}NO_2$, 2315, 2316.
$C_{10}H_{16}N_2$, 4720, 6846, 6350
$C_{10}H_{16}N_2O_3$, 1017.
$C_{10}H_{16}N_2O_3S$, B1841M.
$C_{10}H_{16}N_2O_8$, 8854.
$C_{10}H_{16}O$, 2309, 2609, 2614, 2788, 2789, 3965, 6992, 7424, 8284.
$C_{10}H_{16}O_3$, 4172.
$C_{10}H_{16}O_4$, 2319, 2320, 2321, 5003, 5429.
$C_{10}H_{16}O_5$, 2736, 8028.
$C_{10}H_{16}O_6$, 4192.
$C_{10}H_{17}Cl$, 1960, 4949, 5844.
$C_{10}H_{17}NO$, 2305, 2310,
$C_{10}H_{18}$, 2297, 2612, 3094M, 3131, 3131H, 5002, 5520, 5847, 6238F, 6238M.
$C_{10}H_{18}O$, 1951, 1953, 1955, 2610, 2734, 2735, 2796, 2797, 4173, 5051, 5387, 5532, 6122M, 8135.
$C_{10}H_{18}O_2$, 2304, 6093.
$C_{10}H_{18}O_3$, 195, 8822.
$C_{10}H_{18}O_4$, 412, 4296, 5450, 5455, 5461, 6346, 6349, 7875
$C_{10}H_{18}O_5$, 5416.
$C_{10}H_{18}O_6$, 8102.
$C_{10}H_{19}ClO$, 2379.
$C_{10}H_{19}N$, 1950, 2327, 2344.
$C_{10}H_{20}$, 3117, 5513, B3117M, B3117N.
$C_{10}H_{20}ClNO$, 5379.
$C_{10}H_{20}N_2O_4$, 413.
$C_{10}H_{20}N_2S_4$, 3356.
$C_{10}H_{20}N_2S_4Zn$, 2409.
$C_{10}H_{20}O$, 2332, 2613, 2798, 2799, 3113, 3114, 3115, 5529, 7791.
$C_{10}H_{20}O_2$, 2335, 2352, 2382, 4597, 5146, 6266, 6450, 6543, 8796.
$C_{10}H_{20}O_2\cdot H_2O$, 8138.
$C_{10}H_{20}O_4$, 3236.
$C_{10}H_{21}I$, 3101.
$C_{10}H_{21}N$, 3038, 7015M.
$C_{10}H_{21}NO$, 2333, 2334.
$C_{10}H_{21}NO_2$, 3110.
$C_{10}H_{21}NO_3$, 3109.
$C_{10}H_{22}$, 3099, 6252.
$C_{10}H_{22}N_2O_2$, 7289
$C_{10}H_{22}N_2O_4$, 5289.
$C_{10}H_{22}O$, 545, 3107, 3112, 3734, 4488, 4598, 4600, 4937, 6184, 6270, 6273.
$C_{10}H_{22}O_2$, 3103
$C_{10}H_{22}O_3$, 6332, 6335.
$C_{10}H_{22}O_4S$, 556.
$C_{10}H_{22}O_5$, 8154T.
$C_{10}H_{22}S$, 4944, 8055, B3104M.
$C_{10}H_{22}S_2$, 4931.
$C_{10}H_{23}N$, 3125, 3164, 3264.
$C_{10}H_{23}NO_2$, 8598.
$C_{10}H_{24}O_3S$, 4943.

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$C_{11}H_6O_{10}$, 1365.
$C_{11}H_7N$, 6020, 6021.
$C_{11}H_7NO_2$, 5947.
$C_{11}H_8O$, 5829, 5831.
$C_{11}H_8O_2$, 5828, 5830, 5832, 5941, 5948, B6038R.
$C_{11}H_8O_3$, 5942, 5943, 5944, 5945, 5946, 5949, 5950, 5951, 5952, 6952M.
$C_{11}H_9BrO$, 5989
$C_{11}H_9Cl$, B5844M.
$C_{11}H_9N$, 7488, 7489, 7490.
$C_{11}H_9NO$, 5930, 5931.
$C_{11}H_{10}$, 5877, 5878.
$C_{11}H_{10}N_2O$, 4128.
$C_{11}H_{10}N_2S$, 8249.
$C_{11}H_{10}O$, 3760, 3761, 5993.
$C_{11}H_{10}O_2$, 7264M.
$C_{11}H_{10}S$, 8234.
$C_{11}H_{11}N$, 6050, 6064, 7670, 7671, 7672, 7673, 7674, 7675.
$C_{11}H_{11}NO$, 2592.
$C_{11}H_{11}NO_2$, 2769, 2771, 2774.
$C_{11}H_{11}N_3O_2S$, 8050M.
$C_{11}H_{12}ClNO_3$, 4688.
$C_{11}H_{12}N_2O$, 861, 8848.
$C_{11}H_{12}N_2O_2$, 8687, 8688, 8689.
$C_{11}H_{12}N_2O_4$, 1540M, B1540F.
$C_{11}H_{12}N_2S$, 8267.
$C_{11}H_{12}O$, 5199.
$C_{11}H_{12}O_2$, 2137M, 2744, 2760, 8828.
$C_{11}H_{12}O_3$, 117, 5822.
$C_{11}H_{12}O_4$, 1075, 3951.
$C_{11}H_{13}NO_2$, 4787.
$C_{11}H_{13}NO_3$, 4686.
$C_{11}H_{13}NO_3S$, 4687.
$C_{11}H_{13}N_3O_3$, 8352.
$C_{11}H_{14}N_2\cdot\frac{1}{2}H_2O$, 5001.
$C_{11}H_{14}N_2O$, 3089.
$C_{11}H_{14}O$, 5159, 8827.
$C_{11}H_{14}O_2$, 1453, 1458, 2224, 4767, 8851, 8852.
$C_{11}H_{14}O_3$, 4101, 5478, 7835, 8294.
$C_{11}H_{15}NO$, 8793R.
$C_{11}H_{15}NO_2$, 2144, 2431.
$C_{11}H_{16}$, 1122, 1124, 1212, 1213, 1218, 1241, 1252, 1263, 8349, 8350, 8351, 8369.
$C_{11}H_{16}N_2O_3$, 6976.
$C_{11}H_{16}O$, 2086, 3662, 3663, 3684, 3685, 3686, 3751, 6694, 6695, 6771, 6790, 7180.
$C_{11}H_{16}O_2$, 6691, 6692, 6693, 7750, 7764.
$C_{11}H_{16}O_4$, 4147K.
$C_{11}H_{17}ClN_2O_2$, 6977.
$C_{11}H_{17}N$, 647, 675, 8488, 8512.
$C_{11}H_{17}NO_3$, 5549.
$C_{11}H_{18}N_2O_3$, 6978.
$C_{11}H_{18}N_2$, 4719.

PLAINTIFF'S EXHIBIT 3

MELTING POINT INDEX OF ORGANIC COMPOUNDS

The following table lists the melting points of organic compounds in ascending order of temperature in degrees Centigrade. The compounds are identified by the numbers as given in the table Physical Constants of Organic Compounds.

The letter S preceding any compound number indicates that it is to be found in the **Supplement** to the table.

Melting points will be quoted as they occur in this table. The order of compounds listed with the same melting point is not significant and will be arbitrarily given in the order of the compound numbers. Owing to lack of agreement in values reported by various observers more than one value is sometimes given or the melting point is indicated by a temperature range. In such cases the position in the list is determined by the lower temperature of the stated range or by the *first* of two stated values, the second value being given in parentheses. Where the values are separated by more than 5° C the melting points are listed separately.

Melting points are given in bold face followed by the numbers of the compounds.

Abbreviations: a = anhydrous; h = hydrate.

(**−218**), 2583
−207, 2583
−189.9, 7073
−185.2, 7217
−184, 2587M, 8584
−181, 5006M
−172, 3488
−169.4, 3809
−161.5, 1947
−160.5, 2028
−159.7, 8861
−159, 7359
−156.6, 7805
−151, 6175
−150, 4650, 7892
(**−147.2**), 8586
−146, 5004M, 7062, S4623A
−145, 4950, 5613
−142.5, 3862M
−142.4, S3047M
−141, 4541
−139, 6572
−138.7, 3806
−138.5, 5749
−138, 2590, 6564
−137.6, 8800
−136.4, 478
−135.1, 2019
−135, 1998, 2117, S4628A
−133, S4623B
−131.6(**−129.9**), 6481
−131.3, 2167
−131.2, 4958
−130.7, 7164
−130, 2110, 2196
−130, 468
−127, 7331
−126.6, 3062
−126.4, 3002
−126, S6277J
(**−125.6**), 6498
−125, 6493
−124.6, 7219
−124, 2120, 3078, 3624
−123.6, 10
−123.4, 6495
−123.2, 3902a
−123.1(**−121**), 6715
−123.1, 2160
−122.8, 7344
−122, 7358
−121, 3586
−120, 1090M, 5095
−119.4, 476, 4572
−119.1, 4571
−119, 3804
−118.5, 4957
−118, 6876, S6277Q
−117.3, 3785
−117.2, 4923
−117, 5101
−116.6, 7218
−116.3, 3902a
−115.9, 2072
−115.5, 3681
−114.8, 8396
−114.6, 3783
−114, 2589M
−113, S4028B
−112.6, 2266
−112.4, 2106
−112, 321, 8304, S3117M
(**−112**), 3917
−111.9, 4928
−111.6, 7163
−111.3(**−107**), 2885
−111.2, 3899
−110.8, 8380
−110, 7343, 8829
−108.6, 2565
−108.5(**−106** to **−111**), 3912
−108, 4951, S6277K
−107, 8557M
−105, 534
−104.8, 5628, 8070
−104.7, 7372
−104.5, 2157
−104, 2182
(**−104**), 6876, 7319
−103.7, 3029
−103.5, 2181, 3601
−102.1, 3930
−102, 3917
−101.6, 1276
−101.4, 7362
−101.2, 8099
−101, 6805
−100.6, 3530M
−100.2, 8107W
<**−100**, 2093, 2347
−100, 5124
−99.3, 488, 5145
−99, 537, 2204, 4031
−98.9, 100
−98.5, 4618
−98.2, 2018
−98.1, 103
−98, 2082
−97.8, 5719
−97.6, 5738
−96.9, 2925, 8356
−96.7, 3507, 8744
−96.5, 319
−96, 3276, 8826
−95.8, 4028
−95.2(**−98**), 2178
−95.2, 2235
−95, 213, 6594, 8336
<**−95**, 2232
−94.5, 6498
−94.4, 5039
−94.3, 4550
−94, 7324, S6277R
−93.9(**−92.8**), 1210
−93.66, 5734
−93.5, 4966, 8660
−93.3, 2223, 3046, 3038
−93, 4597
−92.9, 4034
−92.8, 8797
−92.5, 107, 8729
−92, 2100, 3995
−91.9, 7319
−91.5, 2224, 8790
−91.3, 4543
−91.2, 8798
−91, 4566, 8801
−90.6, 8106
−90.5, 4458
−90, 4020, 5000
−89.55, 7280
−89.2 to **−88.6**, 2149
−89, 2150, 2272, 5100
−88.5 to **−88.5**, 5096
−88.2, 4981
−88, 835
−87.7, 378

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283(**300d**), 7848
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283-4d, 432, 5066
285-6, 8687
286, 1482, S7982M
286, 5676, 5782, 8124
286-8d, 4654
286, 794, 2782, 2773, 5502, S4678J
287, 4064
287-8, 4655
288, 5090
288d, 8050
288d, 445
289, 442, 806, 810, 2670, 8788
(**289**), 5341
289-1, 7696R
(**291**), 1431
291.5d, 8806
292, 809
292d, 5065
(**292d**), 8831
293, 1259
293d, 1792, 8833
(**293-5**), 4064
293-5(**280d**), 8689
293, 426, 2862, 5342
293d, 433, 8694
295-300d, 3972, 8982
296, 935
297, 2997
297d, 430
298d, 1479, 5801, 8831
299, 1535, 3000, 4166
>**299**, 1592, 6357
300-305d, S3382M
301d, 6212, 6318
301-3d, 8136a
301-3, 6628.
302, 796, 839
303, 805
305, 448, 1433
306, 1224
307-8, 5161
308-10d, 949
310, 1914a, 3618
310d, 768, 7617
310(**300**), 5089
310-4, 8693
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312-6d, 3988
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315d, 8832
316, 8692
317, 5075
318-20d, 429
319, 808
(**320**), 1005
>**320**, 812, 3133
323, 3291
327d, 6211, 7418
327-8, 1419
328-9d, 8109
330, 767, 4046, 8084
>**330**, 814, S2208M
330, ca., 963
331, 1418
332d, 5341
333d, 4391R
337, 8172
338, 8707
338-40, 7696P
340, 982
345d, 1005
350, 8621
(>**350**), 1419
350d, 1229
354, 377
355d, 2756
360, 3968
>**360**, 764, 2965, 3978
360d, 4404
>**360d**, 8782
364, 6960
365, 407
368, 791
370d, 8783
(**378-80**), 8621
384-6, 826
393d, 4874
400d, 8781
419d, 6371
437-40, S2837J
450d, 1377
470-500d, 4865

PLAINTIFF'S EXHIBIT 3

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PHYSICAL CONSTANTS OF ORGANIC COMPOUNDS (Continued)

No.	Name	Synonyms	Formula	Mol. Wt.	Crystalline form, color and index of refraction	Density g/ml	Melting point, °C	Boiling point, °C	Solubility in grams per 100 ml of: Water	Alcohol	Ether, etc.
7054	**Prehnitic acid**	1,2,3,5-benzenetetracarboxylic acid*	$C_6H_2(COOH)_4$	254.16	pr. (+2H$_2$O) f. w.	....	237 d.	d.	s.	......	s. eth.
7055	**Prehnitylic acid**	2,3,4-trimethylbenzoic acid	$(CH_3)_3C_6H_2COOH$	164.20	pr. f. al	....	167.5	....	s.	s.	s. eth.
7056	**Procaine, hydrochloride**	β-diethylaminoethyl p-aminobenzoate hydrochloride; novocain; ethocain	$NH_2C_6H_4COOC_2H_4N(C_2H_5)_2 \cdot HCl$	272.77	col. need. f. al	0.707^{15}	156	....	160^{20}	3.3^{6}	v. sl. s. eth.; sl. s. chl.
7057	**Proline, hydroxy-L-**	....	$C_4H_7N(OH)COOH$	131.13	rhomb. pl. or fine need.	....	274; (236–41) d	....	25^{0}	v. sl. s.	i. eth.
7058	**Proline, 4-hydroxy-D-**	4-hydroxy-2-pyrrolidinecarboxylic acid	$C_4H_7N(OH)COOH$	131.13	....	....	(a) 274; (b) 238–41 d.	....	....	....	....
7059	**DL-Proline**	dl-2-pyrrolidinecarboxylic acid	$C_4H_8N \cdot COOH$	115.13	....	....	205 d.	....	s.	s.	i. eth.; sl. s. chl., acet., bz.
7060	**D-Proline**	d-2-pyrrolidinecarboxylic acid	C_4H_8NCOOH	115.13	pr.	....	215–20 d	....	....	....	....
7061	**L-Proline**	l-2-pyrrolidinecarboxylic acid	$C_4H_8N \cdot COOH$	115.13	flat need. f. al.; pr. f. w.	....	220–2 d.	....	v. s.	1.55^{19}	i. eth., butyl al., propyl al.
7061M	**Prontosil album.**	See *Sulfanilamide.*									
7062	**Propadiene***	allene; dimethylenemethane	$CH_2:C:CH_2$	40.06	gas	1.787 g/l	−146	−32	....	....	....
7063	**——, dioxo-.**	See *Carbon suboxide.*									
7064	**——, tetraphenyl-**	tetraphenylallene	$(C_6H_5)_2C:C:C(C_6H_5)_2$	344.43	need. or pr. f. dil. al. or acet.	....	166	....	i.	sl. s. c.	s. eth.
7065	**Propanal*.**	See *Propionaldehyde.*									
7066	**——, 2,2-dimethyl-*.**	See *Pivaldehyde.*									
7067	**——, 2-methyl-*.**	See *Isobutyraldehyde.*									
7068	**——, 2-oxo-*, 1-oxime.**	See *Pyruvaldehyde, aldoxime.*									
7069	**——, 3-phenyl-.**	See *Hydrocinnamaldehyde.*									
7070	**Propanamide*.**	See *Propionamide.*									
7071	**——, 2-hydroxy-*.**	See *Lactamide.*									
7072	**——, 2-methyl-*.**	See *Isobutyramide.*									
7073	**Propane***	dimethylmethane	$CH_3CH_2CH_3$	44.09	col. gas	liq. $0.5853\frac{-44.5}{4}$ 2.0096 g/l	−189.9; fr. −187.1	−42.17	6.5^{18} cm³	790^{17} cm³	926^{17} cm³ eth.
7074	**——, 1-amino-2,2-dimethyl-.**	See *Propylamine, β, β-dimethyl-*.*									
7075	**——, 1-amino-2-methyl-.**	See *Isobutylamine.*									
7076	**——, 2,2-bis(ethylsulfonyl)-***	acetone diethylsulfone; sulfonmethane; sulfonal	$(CH_3)_2C(SO_2C_2H_5)_2$	228.32	col. monocl. pr. f. al.	1.260^{11}_{4}	126	300 d	2^{15}; 6.7^{100}	1.2 c., 39 b.	0.84 c. eth.
7077	**——, 1-bromo-*.**	See *Propyl bromide.*									
7078	**——, 2-bromo-*.**	See *Isopropyl bromide.*									
7079	**——, 1-bromo-2-chloro-***	....	$CH_3CHClCH_2Br$	157.45	liq., 1.47449	$1.531\frac{22}{4}$		118.0			....
7080	**——, 2-bromo-1-chloro-***	....	$CH_3CHBrCH_2Cl$	157.45	liq., 1.47763	$1.537\frac{10}{4}$		117.0^{750}			....
7081	**——, 1-bromo-2,2-dimethyl-***	*tert*-butylmethyl bromide	$(CH_3)_3CCH_2Br$	151.06	col. liq.	1.2604^{10}_{0}		$89–91^{710}$	i.	s.	s. eth.
7082	**——, 1-bromo-2-methyl-*.**	See *Isobutyl bromide.*									
7083	**——, 2-bromo-2-methyl-*.**	See *tert-Butyl bromide.*									
7084	**——, 1-chloro-*.**	See *Propyl chloride.*									
7085	**——, 2-chloro-*.**	See *Isopropyl chloride.*									
7086	**——, 1-chloro-2-(β-chloroisopropoxy)-*.**	See *Ether, bis-β-chloroisopropyl.*									
7087	**——, 1-chloro-2,2-dimethyl-***	....	$(CH_3)_3CCH_2Cl$	106.60	....	....		84.4	....	....	....
7088	**——, 1-chloro-2,3-epoxy-*.**	See *Epichlorohydrin.*									
7089	**——, 1-chloro-2-methyl-*.**	See *Isobutyl chloride.*									
7090	**——, 2-chloro-2-methyl-*.**	See *tert-Butyl chloride.*									
7091	**——, 1,1-dibromo-***	propylidene bromide	$CH_3CH_2CHBr_2$	201.91	liq	....	....	ca. 130	....	....	....
7092	**——, 1,2-dibromo-***	propylene bromide; propylene dibromide	$CH_2BrCHBrCH_3$	201.91	col. liq., 1.5203	1.9333^{10}_{4}	−55.5	141.6 (140)	0.25^{20}	s.	v. s. eth.

*Name approved by the International Union of Chemistry.

For explanations and abbreviations see beginning of table.

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PLAINTIFF'S EXHIBIT 4

The Characterization of Organic Compounds
(By Mc Elvain)

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CHAPTER I

Outline of a Procedure for the Characterization of Organic Compounds

When an organic compound is isolated from a natural source or obtained from a reaction *for the first time*, the problem arises of assigning to it a correct structure. This assignment of structure generally requires an expenditure of considerable time and effort because it involves, after purification, the determination of (a) the elemental content, both qualitative and quantitative, (b) the molecular weight, (c) the type and number of functional groups that are present in the molecule and (d) the structure of the carbon framework of the molecule and the manner in which the various functional groups that have been found to be present are attached to this framework.

The structure determination referred to in (d) usually requires the application of degradative experiments — oxidation, hydrolysis, alkali-fusion, etc. — that break down the unknown molecule into smaller fragments of known structure from which it is possible to draw a reasonable conclusion as to the probable structure of the original substance. The final proof of the structure is obtained when a compound of the supposed structure is synthesized by known and unequivocal reactions and when this synthetic compound and the one in question are found to be identical in all of their properties.

Through the prodigious efforts of organic chemists during the past hundred years the structures, together with the physical and chemical properties, of between 300,000 and 400,000 organic compounds have been recorded in the chemical literature. In laboratory work with organic compounds, particularly in research, it frequently is necessary to determine whether or not a certain substance that is encountered is identical with one of the large number of these known compounds or if its structure must be determined by the more involved procedure that is required for a compound

not previously described in the literature. An attempt to locate a single compound with the properties of the one in question among the great number of known organic compounds might, at first glance, have some of the aspects of the search for the needle in the haystack. However, the fortunate fact that this great number of organic compounds can be grouped into relatively few classes has been responsible for the development of systematic procedures of identification that make the characterization of a compound the, properties of which are recorded in the literature a comparatively simple matter.

One characterization procedure which has been found useful and is developed in detail in the subsequent chapters of this book is outlined in the following four steps:

1. The Determination of the Physical Properties and the Establishment of the Purity of the Compound. The melting point of a solid and a combination of the boiling point and the refractive index of a liquid are the most used physical properties for the characterization of organic compounds. The density and the freezing point also are often valuable constants of organic liquids. In the case of an optically active compound the specific rotation is always significant information. The values obtained from the determination of one or more of these physical properties generally furnish a reliable criterion of the purity of an organic compound.

2. Determination of the Class of Compounds, i.e., Aldehydes, Ketones, Acids, Amines, etc., to Which the Compound Belongs. This is done by the systematic application of a series of tests which, as they are applied, are designed to exclude large groups of compounds from consideration and to reduce the number of classes that might contain the compound to be identified until finally only a single class remains. These tests, in the order of their application, are:

(a) *Qualitative analysis* for the elements: nitrogen, halogens and sulfur. Obviously, the presence or absence of any or all of these elements would exclude from consideration certain classes of organic compounds.

(b) Determination of the *solubility* of the compound. Its solubility or insolubility in certain selected solvents makes possible a further reduction in the number of classes that may contain the compound.

PLAINTIFF'S EXHIBIT 6

"Organic Chlorine Compounds"
(By Huntress)

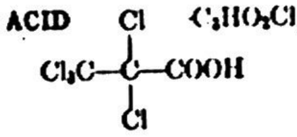
[Ĉ with pyridine + CuO + hydrazine hydrate yields (13) a hexachloronaphthalene ndls. from chlorobenzene, m.p. 202–204° (13) (note that m.p. is close to that of Ĉ).]

Ĉ is stable toward 3% aq. KOH or NaOH for 12 hrs. in cold or even 2 hrs. boilg.; Ĉ is stable to cold 3% alc. KOH or NaOH for 24 hrs., but on htg. for 2 hrs. splits off some 25–30% chlorine and in part resinifies (2).

⊕ Color test with $SbCl_5$ in CCl_4: Ĉ with $SbCl_5$ in CCl_4 gives stable cherry-red color (2). [This response is not given by carbon tetrachloride (3:5100), hexachloroethane (3:4835), hexachlorobenzene (3:4939), or decachloroindane (2).]

3:4886 (1) Ruoff, *Ber.* **9**, 1486–1488 (1876). (2) Shvemberg, Gordon, *J. Gen. Chem.* (*U.S.S.R.*) **2**, 921–926 (1932); *Cent.* **1934**, I 215; *C.A.* **27**, 2439 (1933). (3) Shvemberg, Gordon, *J. Gen. Chem.* (*U.S.S.R.*) **4**, 695–703 (1934); *Cent.* **1935**, II 514; *C.A.* **29**, 2162 (1935). (4) Berthelot, Jungfleisch, *Bull. soc. chim.* (2) **9**, 446–455 (1868); *Ann. chim.* (4) **15**, 330–343 (1868). (5) Shvemberg, Gordon, *J. Gen. Chem.* (*U.S.S.R.*) **4**, 529–551 (1934); *Cent.* **1935**, II 514; *C.A.* **29**, 1804 (1935). (6) Pollak, Gebauer-Fülnegg, Blumenstock-Halward, *Monatsh.* **49**, 199 (1928). (7) Claus, Mielcke, *Ber.* **19**, 1186–1187 (1886). (8) Claus, Wenzlik, *Ber.* **19**, 1169 (1886). (9) Haldin-Davis, *Brit. J. Dermatol. Syphilis* **51**, 380–383 (1939); *C.A.* **34**, 1413 (1940). (10) Heckert (to du Pont Rayon Co.), U.S. 2,060,210, Nov. 10, 1936; *Cent.* **1937**, I 1604; *C.A.* **31**, 555 (1937). (11) Stern, Ger. 411,314, March 26, 1925; *Cent.* **1925**, II 234. (12) B.A.S.F., Ger. 66,611. *Friedländer* **3**, 271. (13) I.G., French 699,492, Feb. 10, 1931; *Cent.* **1931**, 3519.

3:4895 PENTACHLOROPROPIONIC ACID $C_3HO_2Cl_5$ Beil. II — II_1-(112) II_2-(228)



$Cl_3C{-}C(Cl)_2{-}COOH$

M.P. 200–215° (see text (1)).

Colorless cryst. from CCl_4. — Eas. sol. cold aq.

[For prepn. of Ĉ from trichloroacrylic acid (3:1840) with Cl_2 in CCl_4 soln. in sunlight (yield not reported) see (1).]

Ĉ in aq. soln. behaves as a strong acid; on titration it gives a good Neut. Eq., calcd. 246.5; found 245.8 (1).

Ĉ is very unstable: e.g., Ĉ in aq. soln. on warming dec. (1) (with loss of HCl and CO_2 into tetrachloroethylene (3:5460)); presumably this same decompn. occurs on process of taking its m.p., and the value given above refers to rapid htg. on Hg bath.

Salts. The salts of Ĉ can be obtd. by neutralization with metal hydroxide of a cold satd. aq. soln. of Ĉ followed by evapn. to dryness at low temp.; aq. solns. of the salts are also unstable and rapidly decompose yielding metal chloride + CO_2 + tetrachloroethylene (3:5460) (1). [For studies of influence of light on decompn. of these salts see (2) (3) cf. (4).]

[For study of behavior of Ĉ with H_2 + colloidal Pd see (5).]

Acid chloride. Pentachloropropionyl chloride (3:0470) has been obtd. indirectly, i.e., from trichloroacryloyl chloride with Cl_2 in sunlight (6); colorless cryst., m.p. 42°.

3:4895 (1) Böeseken, *Rec. trav. chim.* **46**, 841–843 (1927). (2) Jaeger, *J. Chem. Soc.* **119**, 2070–2076 (1921). (3) Jaeger, Berger, *Rec. trav. chim.* **41**, 72 (1921). (4) Jaeger, *Cent.* **1912**, I 1817–1818; *Cent.* **1911**, II 1851. (5) Böeseken, *Rec. trav. chim.* **35**, 273–274 (1915). (6) Böeseken, Hasselbach, *Rec. trav. chim.* **32**, 11–14 (1913).

II. INDEX OF EMPIRICAL FORMULAS ACCORDING TO PERCENTAGE CHLORINE CONTENT

The empirical formulas for all groups of isomers listed in this book are represented below in diminishing order of their percentage chlorine. Note that there are numerous instances in which this value is the same for several different formulas. An analogous list of empirical formulas arranged according to their molecular weights comprises Index III (page 1383).

To ascertain which specific compounds corresponding to a given formula are described in the text of this volume, see the Empirical Formula Index (page 1359).

% Chlorine	Formula
92.2	CCl_4
89.9	C_2Cl_6
89.1	$CHCl_3$
88.7	C_3Cl_8
87.6	C_2HCl_5
87.0	C_3HCl_7
85.5	C_2Cl_4
85.5	C_3Cl_6
85.0	$C_4H_2Cl_8$
84.8	$C_3H_2Cl_6$
84.7	C_4OCl_{10}
84.5	$C_2H_2Cl_4$
83.5	CH_2Cl_2
82.9	$C_4H_3Cl_7$
82.7	C_3HCl_5
82.5	C_5Cl_8
82.0	$C_3H_3Cl_5$
81.6	C_4Cl_6
81.1	$C_4H_2OCl_8$
81.0	C_2HCl_3
81.0	$C_4H_2Cl_6$
80.3	$C_4H_4Cl_6$
79.7	$C_2H_3Cl_3$
78.8	$C_3H_2Cl_4$
78.3	C_4HCl_5
78.0	C_2OCl_4
78.0	$C_3H_4Cl_4$
77.6	$C_4H_3Cl_5$
77.0	C_3HOCl_5
77.0	$C_4H_5Cl_5$
76.9	C_4OCl_6
75.8	$C_3O_2Cl_6$
74.7	C_6Cl_6
74.7	C_8Cl_8
74.5	C_7HCl_7
73.9	$C_4H_2Cl_4$
73.2	C_3OCl_4
73.1	$C_2H_2Cl_2$
73.1	$C_3H_3Cl_3$
73.1	$C_4H_4Cl_4$
73.1	$C_6H_6Cl_6$
72.5	$C_5H_7Cl_5$
72.4	$C_3H_2OCl_4$
72.4	$C_4H_6Cl_4$
72.2	C_2HOCl_3
72.1	$C_3H_5Cl_3$
72.0	$C_3HO_2Cl_5$
71.7	$COCl_2$
71.7	$C_2O_2Cl_4$
71.7	$C_9H_4Cl_8$
71.7	$C_3O_3Cl_6$
71.7	$C_3H_4OCl_4$
71.2	$C_2H_3OCl_3$
71.2	$C_7H_2Cl_6$
70.8	C_6HCl_5
70.7	C_6OCl_6
70.3	$C_{10}Cl_8$
70.2	CH_3Cl
68.9	$C_4O_3Cl_6$
68.6	$C_{10}H_{10}Cl_8$
68.0	C_7OCl_6
67.6	$C_4H_3Cl_3$
67.6	$C_8H_6Cl_6$
67.2	$C_6O_2Cl_6$
67.1	$C_7H_3Cl_5$
66.9	$C_3H_2O_2Cl_4$
66.7	$C_4H_5Cl_3$
66.6	C_6HOCl_5
65.9	$C_3H_3OCl_3$
65.9	$C_4H_7Cl_3$
65.9	$C_6H_6O_2Cl_6$
65.7	$C_6H_2Cl_4$
65.1	$C_2HO_2Cl_3$
64.5	$C_5H_2OCl_4$
64.5	$C_6H_6Cl_4$
64.3	$C_2H_3O_2Cl_3$
63.9	$C_3H_4Cl_2$
63.9	$C_4O_2Cl_4$
63.7	C_7HOCl_5
63.3	$C_4H_2O_2Cl_4$
63.3	$C_6H_{10}Cl_4$
62.8	$C_2H_2OCl_2$
62.8	$C_3H_6Cl_2$
62.8	$C_4H_4O_2Cl_4$
61.7	$C_2H_4OCl_2$
61.7	$C_7H_4Cl_4$
61.3	$C_5H_7Cl_3$
61.2	$C_6H_2OCl_4$
60.6	$C_3HO_2Cl_3$
60.6	$C_4H_5OCl_3$

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III. INDEX OF EMPIRICAL FORMULAS ACCORDING TO MOLECULAR WEIGHTS

The empirical formulas for all groups of isomers listed in this book are represented below in increasing order of their molecular weights. Note that there are numerous instances in which this value is the same for several different formulas. An analogous list of empirical formulas arranged according to their percentage chlorine comprises Index II (page 1380).

To ascertain which specific compounds corresponding to a given formula are described in the text of this volume see the Empirical Formula Index (page 1359).

M.W.	Formula	M.W.	Formula	M.W.	Formula
50.5	CH_3Cl	113.0	$C_3H_6Cl_2$	136.5	$C_4H_5O_3Cl$
60.5	C_2HCl	115.0	$C_2H_4OCl_2$	136.6	$C_5H_9O_2Cl$
62.5	C_2H_3Cl	116.6	C_6H_9Cl	136.6	$C_6H_{13}OCl$
64.5	C_2H_5Cl	118.6	C_5H_7OCl	136.6	C_8H_5Cl
74.5	C_3H_3Cl	118.6	$C_6H_{11}Cl$	137.0	$C_5H_6Cl_2$
76.5	C_3H_5Cl	119.0	C_4Cl_2	138.6	$C_4H_7O_3Cl$
78.5	C_2H_3OCl	119.4	$CHCl_3$	138.6	C_8H_7Cl
78.5	C_3H_7Cl	120.5	$C_4H_5O_2Cl$	139.0	$C_5H_8Cl_2$
80.5	C_2H_5OCl	120.6	C_5H_9OCl	140.6	C_7H_5OCl
84.9	CH_2Cl_2	120.6	$C_6H_{13}Cl$	140.6	C_8H_9Cl
86.5	C_4H_3Cl	122.6	$C_4H_7O_2Cl$	141.0	$C_3H_2O_2Cl_2$
88.5	C_4H_5Cl	122.6	$C_5H_{11}OCl$	141.0	$C_4H_6OCl_2$
90.5	C_3H_3OCl	123.0	$C_4H_4Cl_2$	141.0	$C_5H_{10}Cl_2$
90.6	C_4H_7Cl	124.1	C_2OCl_3	142.5	$C_6H_3O_2Cl$
92.5	C_3H_5OCl	124.6	$C_4H_9O_2Cl$	142.6	C_7H_7OCl
92.6	C_4H_9Cl	125.0	$C_4H_6Cl_2$	143.0	$C_3H_4O_2Cl_2$
94.5	$C_2H_3O_2Cl$	126.6	C_7H_7Cl	143.0	$C_4H_8OCl_2$
94.5	C_3H_7OCl	126.9	$C_2O_2Cl_2$	144.6	$C_6H_5O_2Cl$
94.9	C_2Cl_2	127.0	$C_3H_4OCl_2$	144.6	$C_8H_{13}Cl$
97.0	$C_2H_2Cl_2$	127.0	$C_4H_8Cl_2$	145.4	$C_3H_3Cl_3$
98.9	$COCl_2$	128.6	C_6H_5OCl	146.6	$C_7H_{11}OCl$
99.0	$C_2H_4Cl_2$	128.6	C_7H_9Cl	146.7	$C_8H_{15}Cl$
102.6	C_5H_7Cl	128.9	$C_2H_2O_2Cl_2$	147.0	$C_6H_4Cl_2$
104.5	C_3HO_2Cl	129.0	$C_3H_6OCl_2$	147.4	C_2HOCl_3
104.5	C_4H_5OCl	130.5	$C_5H_3O_2Cl$	147.4	$C_3H_5Cl_3$
104.6	C_5H_9Cl	130.6	$C_7H_{11}Cl$	148.6	$C_6H_9O_2Cl$
106.5	$C_3H_3O_2Cl$	131.0	$C_2H_4O_2Cl_2$	148.6	$C_7H_{13}OCl$
106.6	C_4H_7OCl	131.4	C_2HCl_3	148.7	$C_8H_{17}Cl$
106.6	$C_5H_{11}Cl$	132.5	C_4HO_3Cl	149.0	$C_6H_6Cl_2$
108.5	$C_3H_5O_2Cl$	132.6	C_6H_9OCl	149.4	$C_2H_3OCl_3$
108.6	C_4H_9OCl	133.6	$C_7H_{13}Cl$	150.5	$C_4H_3O_4Cl$
110.5	$C_3H_7O_2Cl$	133.4	$C_2H_3Cl_3$	150.6	$C_5H_7O_3Cl$
111.0	$C_3H_4Cl_2$	134.6	$C_5H_7O_2Cl$	150.6	$C_6H_{11}O_2Cl$
112.6	C_6H_5Cl	134.6	$C_6H_{11}OCl$	150.7	$C_7H_{15}OCl$
112.9	$C_2H_2OCl_2$	134.7	$C_7H_{15}Cl$	151.0	$C_6H_8Cl_2$

M.W.	Formula	M.W.	Formula	M.W.	Formula
218.8	$C_{12}H_{23}OCl$	244.4	$C_5H_7Cl_5$	285.6	$C_{14}H_{11}Cl_3$
218.8	$C_{13}H_{27}Cl$	244.6	$C_9H_5O_6Cl$	285.9	$C_8O_3Cl_4$
219.5	$C_6H_9O_2Cl_3$	244.7	$C_{14}H_9O_2Cl$	288.9	$C_{17}H_{33}OCl$
219.9	$C_6H_6Cl_4$	245.9	$C_6O_2Cl_4$	288.9	$C_{18}H_{37}Cl$
220.8	$C_{12}H_{25}OCl$	246.3	$C_3HO_2Cl_5$	290.8	$C_6H_6Cl_6$
221.0	$C_8H_6O_3Cl_2$	246.7	$C_{14}H_{11}O_2Cl$	290.8	$C_{20}H_{15}Cl$
221.5	$C_5H_7O_3Cl_3$	246.8	$C_{14}H_{27}OCl$	294.4	$C_7HO_2Cl_5$
221.5	$C_6H_{11}O_2Cl_3$	246.9	$C_{15}H_{31}Cl$	296.8	$C_3O_3Cl_6$
221.9	$C_4O_2Cl_4$	247.1	$C_{14}H_8Cl_2$	298.8	$C_7H_2Cl_6$
223.1	$C_{12}H_8Cl_2$	247.9	$C_6H_2O_2Cl_4$	299.3	$C_4H_3Cl_7$
223.5	$C_8H_5OCl_3$	248.8	C_3Cl_6	300.8	C_6OCl_6
223.9	$C_4H_2O_2Cl_4$	248.8	$C_{14}H_{29}OCl$	300.9	$C_{18}H_{33}OCl$
224.0	$C_6H_{10}Cl_4$	249.1	$C_{14}H_{10}Cl_2$	302.9	$C_{18}H_{35}OCl$
225.1	$C_9H_{14}O_2Cl_2$	250.4	C_6HCl_5	303.9	$C_8H_2O_4Cl_4$
225.4	$C_7H_3O_2Cl_3$	250.8	$C_3H_2Cl_6$	304.9	$C_{18}H_{37}OCl$
225.5	$C_8H_7OCl_3$	251.1	$C_{13}H_8OCl_2$	308.8	$C_4O_3Cl_6$
225.9	$C_4H_4O_2Cl_4$	251.2	$C_{14}H_{12}Cl_2$	312.8	C_7OCl_6
226.3	C_4HCl_5	255.5	$C_8H_5O_3Cl_3$	316.8	$C_6O_2Cl_6$
226.7	$C_{12}H_{15}O_2Cl$	258.4	$C_6H_9Cl_5$	318.0	$C_{14}H_8Cl_4$
228.4	$C_4H_3Cl_5$	259.9	$C_7H_2O_2Cl_4$	319.7	C_3Cl_8
229.5	$C_6H_3O_3Cl_3$	260.8	C_4Cl_6	319.7	$C_{12}H_{21}O_3Cl_3$
230.0	$C_7H_4Cl_4$	260.8	$C_{15}H_{29}OCl$	320.1	$C_{14}H_{10}Cl_4$
230.3	C_3HOCl_5	260.9	$C_{16}H_{33}Cl$	322.8	$C_6H_6O_2Cl_6$
230.4	$C_4H_5Cl_5$	262.8	$C_4H_2Cl_6$	333.3	C_7HCl_7
230.7	$C_{14}H_{11}OCl$	264.4	$C_7H_3Cl_5$	333.8	$C_4H_2Cl_8$
231.5	$C_{10}H_5Cl_3$	264.8	$C_4H_4Cl_6$	343.7	C_5Cl_8
231.9	$C_6H_2OCl_4$	265.1	$C_{14}H_{10}OCl_2$	349.8	$C_4H_2OCl_8$
232.8	$C_{13}H_{25}OCl$	266.4	C_6HOCl_5	354.5	$C_{14}H_9Cl_5$
232.8	$C_{14}H_{29}Cl$	270.0	$C_{10}H_8Cl_4$	388.9	$C_{14}H_8Cl_6$
233.5	$C_7H_{11}O_2Cl_3$	274.9	$C_{16}H_{31}OCl$	403.8	$C_{10}Cl_8$
234.7	$C_{11}H_{19}O_3Cl$	274.9	$C_{17}H_{35}Cl$	418.6	C_4OCl_{10}
235.0	$C_8H_4O_4Cl_2$	276.8	C_4OCl_6		
235.5	$C_6H_9O_3Cl_3$	276.9	$C_{16}H_{33}OCl$		
236.8	C_2Cl_6	278.4	C_7HOCl_5		
237.1	$C_{12}H_{22}Cl_2$	278.8	$C_{18}H_{27}Cl$		
238.1	$C_{10}H_{16}O_2Cl_2$	280.8	$C_3O_2Cl_6$		
239.9	$C_4H_2O_3Cl_4$	281.1	$C_{14}H_{10}O_2Cl_2$		
242.7	$C_{14}H_7O_2Cl$	283.6	$C_{14}H_9Cl_3$		
243.1	$C_9H_{16}O_3Cl_2$	284.8	C_6Cl_6		
243.9	$C_7H_2OCl_4$	285.2	C_3HCl_7		

PLAINTIFF'S EXHIBIT 7

"The Encyclopedia of Chemistry"
(By Clark & Hawley)

and have pH value less than 7. Considered from a broader standpoint, any compound that is a proton donor is an acid. With solvents other than water, the proton donor definition is fruitful. In water solution, however, acids are generally considered to be compounds capable of producing hydrogen or, better, hydronium (H_3O^+ or $H \cdot H_2O^+$) ions.

There are well over 100 kinds of acids that are commercially available. They are prepared in several ways: (1) By the reaction of a salt with an acid of a higher boiling point than the acid sought. For example, sulfuric acid has a high boiling point and it is stable. It reacts with chlorides to liberate hydrogen chloride which has a lower boiling point than sulfuric acid. Hydrogen chloride dissolved in water forms hydrochloric acid:

$$NaCl + H_2SO_4 \rightarrow HCl + NaHSO_4$$

$$HCl + H_2O \rightarrow H_3O^+ + Cl^-.$$

(2) The addition of water to an oxide, usually of a nonmetal (and called an acid anhydride):

$$CO_2 + H_2O \rightarrow H_2CO_3$$

$$SO_3 + H_2O \rightarrow H_2SO_4\,,$$

(3) *Special methods:* Sulfuric acid is made by the chamber and the contact processes. Nitric acid is made by the catalytic oxidation of ammonia in the Ostwald process. Hydrogen chloride can also be made by direct combination of the elements. The gas is subsequently dissolved in water to form hydrochloric acid.

The *mineral acids* include sulfuric (H_2SO_4), hydrochloric (HCl), nitric (HNO_3), and phosphoric (H_3PO_4). Organic acids contain carbon, and usually contain the group or radical —COOH, called the carboxyl radical. Such acids are *carboxylic acids*. Formic acid (HCOOH), acetic acid (CH_3COOH), and propionic acid (C_2H_5COOH) are in this group. Acids with a larger number of carbon atoms such as palmitic ($C_{15}H_{31}COOH$) and stearic ($C_{17}H_{35}COOH$), are acids that may be derived from fats and oils. Hence this entire group of carboxylic acids is called the *fatty acids*. Some monocarboxylic acids may be unsaturated such as acrylic (or propenoic) acid ($CH_2{=}CHCOOH$) and oleic acid ($C_{17}H_{33}COOH$).

Dicarboxylic acids contain two carboxyl groups. The simplest is oxalic acid (COOH·COOH), a white solid, melting point 189°C as the dihydrate [$(COOH)_2 \cdot 2H_2O$]. This compound might also be called ethanedioic acid. Propanedioic acid, better known as malonic acid ($COOHCH_2COOH$), m.p. 135.6°C, is also in this group.

Aromatic carboxylic acids include benzoic acid (C_6H_5COOH), found in cranberries. It is used as a preservative for foods. Its sodium salt, sodium benzoate, is sometimes used to preserve fruit juices and tomato ketchup.

Three phthalic acids are known, of which the ortho- is the most important. It is $C_6H_4(COOH)_2$, 1,2-benzenedicarboxylic acid, m.p. 206–208°C. Its anhydride, phthalic anhydride [$C_6H_4(CO)_2O$], is used to polymerize with glycerol to form a useful resin.

Phenol, sometimes called carbolic acid, (C_6H_5OH), is an organic acid that reacts with sodium hydroxide, a typical base, to form sodium phenoxide (or sodium phenolate) and water. $C_6H_5OH + NaOH \rightarrow C_6H_5ONa + H_2O$. In contrast to carboxylic acids, phenols in general do not react with sodium hydrogen carbonate (bicarbonate of soda) ($NaHCO_3$), but many carboxylic acids do.

Amino acids are a group of 21 nitrogen-containing acids that may be derived from the decomposition of complex proteins. They all contain at least one carboxyl group.

Strong acids are those that are completely dissociated into ions in water solution. Weak acids show small tendency to form ions. For example, the reaction $HCl + H_2O \rightarrow H_3O^+ + Cl^-$ goes practically to completion when hydrochloric acid, a strong acid, is formed. Acetic acid solution $CH_3COOH + H_2O \rightleftharpoons CH_3COO^- + H_3O^+$ forms only a few ions, and its properties are those of a weak acid. In this sense, glacial acetic acid is even weaker than a solution of hydrogen acetate in water. Concentrated sulfuric acid is weak and dilute sulfuric acid is strong.

The dissociation of the first hydronium ion from an acid in general proceeds more readily than a second. For example, $H_3PO_4 + H_2O \rightarrow H_3O^+ + H_2PO_4^-$ is easier than $H_2PO_4^- + H_2O \rightarrow H_3O^+ + HPO_4^{--}$, and far easier than $HPO_4^{--} + H_2O \rightarrow H_3O^+ + PO_4^{---}$.

The dissociation constant (or ionization constant) for an acid is a measure of the extent to which it forms hydrogen (or hydronium) (H^+ or H_3O^+) ions in water solution. In the case of acetic acid

$$\frac{[H^+] \times [CH_3COO^-]}{[CH_3COOH]} = 1.75 \times 10^{-5}$$

This dissociation constant has a larger value for strong acids than for weak ones. In cases in which an acid dissociates more than one hydrogen (hydronium) ion, the value for K_A for the second hydrogen ion is always smaller than that for the first.

ELBERT C. WEAVER

Cross-references: *Acid Number, Amino Acids, Carboxylic Acids, Anhydrides, Acids (Mineral), Fatty Acids, Ions*

ACIDS, MINERAL

Sulfuric Acid

The mineral acids are sulfuric acid, nitric acid, mixed acid, hydrochloric, hydrobromic and hydrofluoric acids, phosphoric acid, and boric acid. They are called mineral acids to indicate that they are derived from minerals, in contrast to the organic acids which are found in vegetable and animal matter.

Sulfuric acid is the most important of the mineral acids, as shown by the production figures. One reason is that it is relatively cheap; the overall figure for sulfuric acid sold in 1952 was about $17 a ton (in terms of 100% H_2SO_4); another is that by adding sulfuric acid to the salts of the

PLAINTIFF'S EXHIBIT 7

terials; and important additions to the knowledge of steelmaking, steel construction, welding, and metalworking. The Institute is active in assisting the coal products industry to map its future and in investigating recovery processes and methods for utilizing coal chemicals. Its work in petroleum chemistry and technology is recognized internationally as authoritative. On other fellowships new chemicals and processes are being evolved, new foods are being studied, novel products from corn and soybeans are being examined for possible commercial applications, textiles and paper are being given wide research. New kinds of plastics, novel organic protective coatings, new types of floor and wall coverings, commodity standards, new silicones, better life preservers, new insecticides, and new medicinal agents are coming from still other fellowships. The work of Mellon Institute has added in many important ways to the progress of American technology and has aided in bringing many of industry's exploits to successful use by the armed forces.

Scientific inquiry of chiefly or entirely a theoretical nature often results as a bypath of applied research. This fact has been brought out repeatedly in publications that have come from programs relating to problems of large scope, such as improved or new industries. Long-range applied investigations conducted in Mellon Institute, especially for bettering or extending sectors of manufacturing, evolve into the acquirement of all pertinent fundamentals and then the systematic employment of these essentials in contriving superior processes and products. Fellowship research of this kind, the very architecture of technology, has yielded by donor permission 2707 contributions to the literature of science, 1910–1955, inclusive, or 75% of the Institute's total publications.

Objectives. From its inception Mellon Institute has maintained three principal objectives: the conduct of productive research in both the pure and applied sciences, the advanced training of research workers in the fields of their specializations, and the dissemination of scientific information of constructive value to the professions concerned and to the general public. Activating the community to a better appreciation of the sciences and their beneficial impact on human relations, pointing out to industry where science and scientists can be of assistance, carrying forward investigations relating to public needs, turning out skilled research men, Mellon Institute has a long record as a creative organization for the service and advantage of humanity.

Mellon Institute as a national science center constantly fosters the friendship of the professions interested in its work. Contacts are maintained with numerous scientists and scientific organizations, many societies meet in the Institute, and this communication adds valuably to the fertility of research in the organization. Of similar essentiality are continuous and helpful public relations.

WILLIAM A. HAMOR

Cross-references: *Chemical Research*

MELTING POINT

The melting point or freezing point of a pure substance is the temperature at which its crystals are in equilibrium with the liquid phase at atmospheric pressure. It is usually called the melting point when the equilibrium temperature is approached by heating the solid. Ordinarily, therefore, melting points refer to temperatures above 0°C, the melting point of ice.

Conversion from the crystalline to the liquid state is invariably accompanied by the absorption of a definite quantity of heat per mole, known as the *molar heat of fusion*. For ice, the heat of fusion is 79.7 calories per gram. Addition of heat to an intimate mixture of ice and water at 0°C will therefore result in the conversion of one gram of ice to water for every 79.7 calories added. This changes the proportion of ice to water but the temperature of the mixture will remain at 0°C as long as any ice remains. Thus for a pure substance melting takes place at constant temperature.

The presence of dissolved impurity lowers the temperature of equilibrium between the crystalline and liquid phase and therefore causes the melting to take place over a temperature range. That is, as the ice crystals are melted, the liquid phase becomes more and more dilute with respect to the impurity so that the equilibrium temperature gradually rises. The temperature at which the last ice crystals disappear is the melting-point of the mixture and is below the melting point of pure ice.

Melting point determinations have been particularly useful to the organic chemist for identification and characterization of compounds and as a criterion of purity. For this purpose sufficiently accurate determinations can usually be made by means of the capillary method, which is simple and rapid and which requires only a few milligrams of the material. A thin-walled glass capillary containing a small amount of the finely divided sample is immersed in a liquid bath the temperature of which is slowly raised at a controlled rate. The melting is observed visually and the corresponding temperature recorded. A sharp melting point usually indicates high purity. The melting point can be used to identify an unknown compound. For corroboration the method of *mixed melting points* is often used. About equal parts of the unknown compound and of the substance having the same melting point, and therefore thought to be the same compound, are intimately mixed and a capillary melting point determination is made on the mixture. If the unknown and the known compounds are not identical, the mixture will melt over a range and will be completely melted at a temperature considerably below the melting point of the original compounds.

Lowering of the melting point by addition of impurities has been used to advantage in metallurgy in developing alloys for specialized uses where the melting point is an important factor. Thus, Wood's metal, which melts at 70°C, is a mixture of 50 parts of bismuth (m.p., 271°C), 25 parts of lead (m.p., 327°C), 12.5 parts of tin (m.p., 231.9°C), and 12.5 parts of cadmium (m.p. 321°C).

PY AVAILABLE
inal bound volume

PLAINTIFF'S EXHIBIT 7

Food Products. One of the economically most important extraction operations is the production of beet sugar, which currently accounts for about one-third of the world's sugar production. By the use of countercurrent extraction, a dark impure solution containing as high as 12% sucrose is produced. This syrup, by successive purifications and concentrations, may be processed to recover as high as 85 or 90% of the sucrose originally present in the beets as a chemically pure product.

An extraction process that has become of considerable importance in this country during the past few years is the manufacturing of soluble coffee products. By countercurrent extraction with hot water, practically all the solubles may be extracted from the roasted and ground coffee berries. This extract is ordinarily of such a concentration that it may be directly dried by drum or spray driers to produce the finished product.

Metals Purification. Solvent extraction has been found to be the most satisfactory method of purifying uranium, thorium, and plutonium for atomic energy purposes. Most of the extraction systems described in the literature have utilized nitric or hydrochloric acid solutions of the metals. The solvents utilized have been ethers or ether-like compounds, acetone or other ketones, and hydrocarbon solutions of organo-phosphates such as tributyl phosphate. The metals are usually recovered by re-extraction into water followed by a precipitation step of some sort. The use of solvent extraction to purify various other metals is possible, but ordinarily it is not economically feasible.

DENNIS D. FOLEY

Cross-references: *Solvents, Solutions, Diffusion*

SOLVENTS

A solvent is a material capable of dissolving another material (solute) to form a homogenous system composed of more than one chemical substance: a solution. A solvent may be in any physical state; however, the most common type solution is the result of a solvent in a liquid state and a solute in a solid or liquid state.

Solvents are divided into two classifications based on polarity. Polar solvents (alcohols, ketones, etc.) have high dielectric constants. Nonpolar solvents (hydrocarbons) have low dielectric constants.

There are many types of solvents, based on the chemical composition. These general types and typical examples of each are listed below.

Water: Water alone or aqueous solutions of alcohols, salts or other miscible materials.

Hydrocarbons: Naphtha fractions such as V. M. & P Naphtha or Mineral spirits. Aromatic materials such as benzene, toluene or xylene.

Alcohols: Methyl alcohol, Ethyl alcohol, Ethylene glycol.

Ethers: Dimethyl ether, Ethylene glycol monoethyl ether.

Ketones: Acetone, Methyl ethyl ketone, Methyl isobutyl ketone.

Esters: Ethyl acetate, Butyl acetate, Butyl lactate.

Chlorinated Solvents: Chloroform, tetrachloroethane, monochlorobenzene.

Nitrated Solvents: Nitromethane, Nitroethane, 1-Nitropropane, 2-Nitropropane, Nitrobenzene.

Amines: Monoethanolamine, diethanolamine, pyridine.

Liqefied Gases: Ammonia, sulfur dioxide.

The principal solvent-consuming products include paints, varnishes, lacquers and inks; textiles; and pharmaceuticals. Paints, lacquers and inks are the largest, in view of the volume of paint needed for exterior and interior coatings, lacquers and enamels for automobiles and furniture; and the ink used in the vast printing industry.

The solvent power of a solvent for any given solute is the best criterion for determining its value in a process, assuming that other properties such as evaporation rate, stability, odor, purity or toxicity do not make it unsuitable.

The solvent power of aromatic hydrocarbons is usually greater than that of aliphatic hydrocarbons. There are numerous grades of hydrocarbons available, based upon their proportions of aromatic and aliphatic materials. The kauri-butanol test is used as an arbitrary rating of aromatic content. Kauri gum is readily soluble in butyl alcohol but insoluble in hydrocarbons. The kauri-butanol value is a measure of the volume of solvent required to produce turbidity when added to a standard solution containing kauri gum dissolved in butyl alcohol. Naphtha fractions have a kauri-butanol value of about 30, while toluene has 105. The aniline point and mixed aniline point methods are also used to determine the proportion of aromatic material in hydrocarbon solvents. This procedure is based on the fact that at ordinary temperatures, aniline is a poor solvent for aliphatic hydrocarbons and a very good one for aromatic hydrocarbons. The aniline test is conducted by mixing equal volumes of aniline and the solvent to be tested, heating until the solution is absolutely clear and then cooling gradually until the solution clouds. The temperature at which the cloud occurs is the critical solution temperature and is recorded as the aniline point.

Most industries have developed methods of evaluating solvent power for their own particular purpose; however, the dilution ratio test used by the nitrocellulose lacquer industry is used universally. The true (active) solvents for nitrocellulose are esters, ketones, glycol ethers and nitroparaffins, all of which are more expensive than hydrocarbon solvents. Hydrocarbons, in proper proportions, may be added to solutions of nitrocellulose in active solvents without deleterious effect. Excessive quantities of hydrocarbons will cause the nitrocellulose to gel and ultimately precipitate from the solution. For reasons of economics the highest practical proportion of hydrocarbon must be used and the dilution ratio test is the basis for determining this quantity.

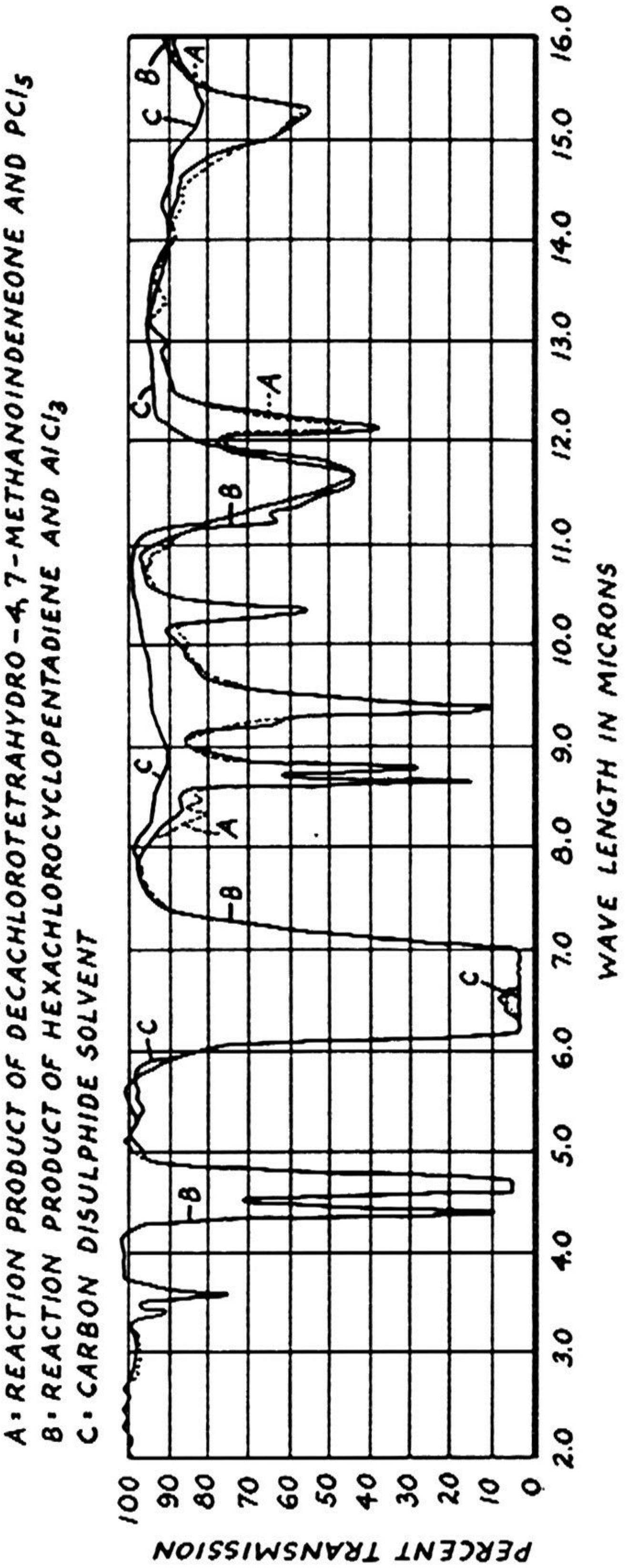
Dilution ratio (sometimes called hydrocarbon tolerance) is the maximum number of unit volumes of hydrocarbon that can be added per unit volume of active solvent to cause the first trace of gelation to occur when the concentration of nitrocellulose in the solution is 8 grams per 100 milliliters. This

PLAINTIFF'S EXHIBIT 8

Feb. 15, 1955 E. E. GILBERT **2,702,305**

PROCESS FOR PREPARING DODECACHLOROTETRAHYDRO-4,7-METHANOINDENE

Filed April 6, 1951



SPECTROGRAM OF DODECACHLOROTETRAHYDRO-4,7-METHANOINDENE

INVENTOR.
EVERETT E. GILBERT
BY Elizabeth Hunter
ATTORNEY.

United States Patent Office

2,702,305

Patented Feb. 15, 1955

2,702,305

PROCESS FOR PREPARING DODECACHLOROTETRAHYDRO-4,7-METHANOINDENE

Everett E. Gilbert, Flushing, N. Y., assignor to Allied Chemical & Dye Corporation, New York, N. Y., a corporation of New York

Application April 6, 1951, Serial No. 219,739

3 Claims. (Cl. 260—648)

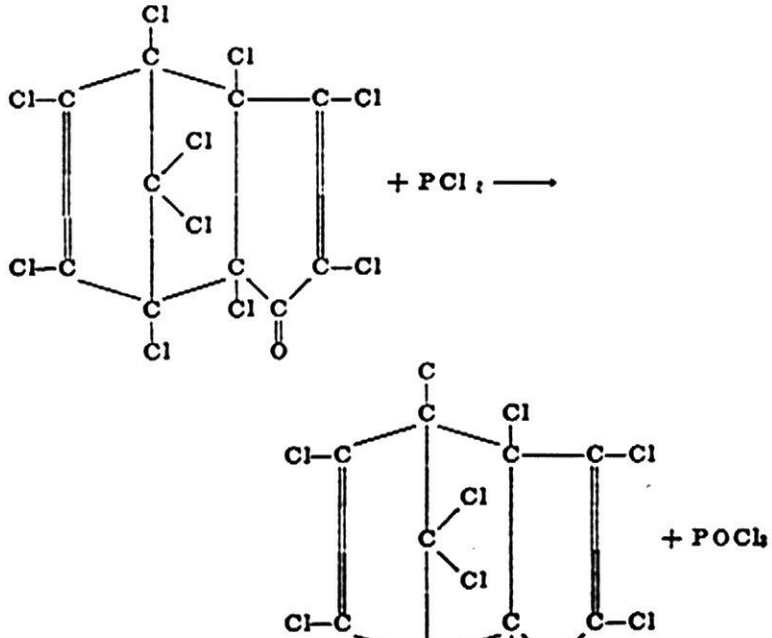
This invention relates to a new method for preparing dodecachlorotetrahydro-4,7-methanoindene, a dimer of hexachlorocyclopentadiene, by the reaction of phosphorus pentachloride with decachlorotetrahydro-4,7-methanoindeneone.

Hexachlorocyclopentadiene dimer has been prepared in the past by the condensation of hexachlorocyclopentadiene with aluminum chloride as described in J. Am.. Chem. Soc. 71, page 954, March 1949, in the description of which process no structure was assigned to the hexachlorocyclopentadiene dimer.

I have now discovered the new process described below for preparing hexachlorocyclopentadiene dimer (dodecachlorotetrahydro-4,7-methanoindene). This compound is useful as an intermediate in carrying out chemical reactions and as an insecticide.

The decachlorotetrahydro - 4,7 - methanoindeneone which forms the starting material for my new reaction may be prepared, for example, as described in my copending application with Silvio L. Giolito, Serial No. 196,123, filed November 17, 1950 now U. S. Patent 2,616,928, by condensing two molecules of hexachlorocyclopentadiene with the aid of sulfur trioxide to form a hexachlorocyclopentadiene-SO_3 reaction product and hydrolyzing the resulting reaction product to the ketone. It is believed to be the 2,3,3*a*,4,5,6,7,7*a*,8,8-decachloro-3*a*,4,7,7*a*-tetrahydro-4,7-methanoindene-1-one illustrated below, and usually exists in the form of a hydrate in a wide range of degrees of hydration.

The reaction as carried out according to my invention proceeds as follows:



In the drawing, the single figure illustrates the infrared spectrogram of dodecachlorotetrahydro-4,7-methanoindene. The dotted line A illustrates the spectrogram of the dimer as prepared according to the process of my invention; the solid line B illustrates the spectrogram of the dimer resulting as the reaction product of hexachlorocyclopentadiene with aluminum chloride. These spectrograms are substantially identical. Solid line C is the spectrogram of carbon disulfide used as solvent in preparing the spectrograms.

In carrying out the process of my invention, decachlorotetrahydro - 4,7 - methanoindeneone, preferably in substantially anhydrous form or in as low a state of hydration as practicable, is reacted with phosphorus pentachloride to effect the replacement of the ketone group with chlorine, for example by mixing the reactants and heating the mixture. The resulting oily reaction product is then cooled and drowned in water, whereupon solid hexachlorocyclopentadiene dimer crystallizes and may be recovered as by filtration and, if desired, washed with a liquid such as methanol which is a solvent for decachlorotetrahydro-4,7-methanoindeneone, but a nonsolvent for the reaction product, to dissolve any unreacted decachlorotetrahydro-4,7-methanoindeneone which may be present. The crude dimer product may then be crystallized from a suitable solvent if desired, such as benzene, isopropanol or the like, and if desired may be further purified of residual decachlorotetrahydro-4,7-methanoindeneone, by first dissolving it in a hot hydrocarbon solvent such as benzene and then precipitating it out of solution by the addition of a hydrocarbon-miscible nonsolvent for the dodecachlorotetrahydro-4,7-methanoindene, such as methanol, in which the decachlorotetrahydro-4,7-methanoindeneone is soluble. The product may be further purified if desired, by recrystallization from an aliphatic alcohol such as isopropanol. The resulting dodecachlorotetrahydro-4,7-methanoindene is a white, crystalline solid which sublimes without melting at temperatures above about 240° C., is appreciably soluble in benzene, acetone, kerosene, carbon tetrachloride, etc. and while virtually insoluble in methanol, is appreciably soluble in the lower aliphatic alcohols having two or more carbon atoms, such as ethanol, isopropanol, etc., particularly at elevated temperatures.

The quantity of PCl_5 to be used in the preparation of the dimer is not critical, but for best yields should be at least the molecular equivalent of the decachlorotetrahydro-4,7-methanoindeneone, so as to furnish two chlorine atoms to replace the keto oxygen as indicated in the reaction above illustrated, and to react with any water of hydration present. Thus, although it is desirable to use decachlorotetrahydro-4,7-methanoindeneone starting material in as low a state of hydration as possible, nevertheless, hydrated forms may be used if a sufficient quantity of PCl_5 is employed to react with all the water of hydration as well as with the keto oxygen, forming $POCl_3$ and HCl, and when the expression "equimolecular quantities" is used in the claims to describe the proportions of reactants, it is to be understood to mean a quantity of PCl_5 sufficient to react with the keto oxygen and also with the water of hydration present. However, as the use of excesses of PCl_5 are not only wasteful, but entail handling of the troublesome HCl formed as a byproduct of the reaction of PCl_5 with the water of hydration, it is preferable to dehydrate the decachlorotetrahydro-4,7-methanoindeneone as much as practicable before carrying out the reaction. In general, therefore, quantities of PCl_5 only slightly in excess of the molecular equivalent of the decachlorotetrahydro-4,7-methanoindeneone as defined are desirable.

The temperature to which the mixture of decachlorotetrahydro-4,7-methanoindeneone and PCl_5 is heated should be at least about 70° C., preferably between about 115° C. and about 160° C. The reaction is usually complete in a period of between about one hour and about six hours. The desired reaction product is separated from the resulting oily reaction mixture, for example by cooling and drowning in an excess of water, for example between about 5 and about 10 volumes of water per volume of the oily reaction mixture, whereupon solid dodecachlorotetrahydro-4,7-methanoindene crystallizes and may be separated by suitable means such as filtration, etc. The resulting crude product is purified by washing with a solvent for decachlorotetrahydro-4,7-methanoindeneone, for example methanol.

The hexachlorocyclopentadiene dimer prepared according to my invention, has the infrared spectrogram shown as dotted line A in the figure. It is identical with the spectrogram of the dimer of hexachlorocyclopentadiene prepared by reacting hexachlorocyclopentadiene with aluminum chloride as described in J. Am. Chem.

Soc. 71, page 954, March 1949, as indicated by the infrared spectrogram of a material made by the $AlCl_3$ condensation method which is shown as solid line B in the figure.

The infrared spectrograms shown in the figure were prepared on a standard infrared recording spectrophotometer designed for measuring and recording the infrared transmission of solids, liquids and gases, comprising a double infrared beam which scans the spectrum through the wave length range 2.0 to 16 microns, one part of the beam passing through the sample under study, the other passing through a compensating cell. If the sample under study absorbs radiation, the two beams become unequal. The magnitude of this inequality is a measure of the transmission of the sample of the particular wave length, and the record of these differences within the range of wave lengths scanned is the infrared spectrogram, recorded as an ink drawn line on a chart graduated in percent transmission as ordinates and in wave length as abscissae.

Solid samples, such as the compound of my invention, are conveniently measured in solution. The spectrogram shown in the figures was measured by dissolving 0.5 gram of the solid in carbon disulfide and diluting to 10 ml. with the solvent. A small amount of the solution was then introduced into a liquid cell with sodium chloride windows and sealed. The cell was placed in the spectrophotometer in the path of one of the beams as described above.

The infrared spectrogram of any chemical compound serves as an accurate means for identifying the compound. It has been compared with a human fingerprint in its ability to identify a compound with certainty. The characteristic reproducibility of the infrared spectrogram of a given compound is due to the facts that when a molecule is excited by infrared radiation it absorbs energy to a greater degree at some wave length than at others, and that the amount of absorption depends on the configuration and upon the linkages of the atoms composing the molecule. Accordingly, this spectrogram identifies and characterizes the hexachlorocyclopentadiene dimer with certainty.

The following specific example further illustrates my invention.

Example

Five parts of hydrated decachlorotetrahydro-4,7-methanoindeneone, purified by solution in methanol, precipitation with water and drying, were mixed with 21 parts of phosphorus pentachloride and the mixture heated for three hours at 125°–150° C. The resulting reaction mixture was cooled to 20° C. and drowned in water. The solid material which formed was filtered and water washed, then washed with methanol to dissolve any unreacted decachlorotetrahydromethanoindeneone which might be present, and dissolved in hot benzene. The benzene solution was mixed with a large excess of methanol to reprecipitate the product, which was filtered, dried and then recrystallized from isopropanol.

Two parts of hexachlorocyclopentadiene dimer were obtained. An infrared spectrogram prepared from this product was found to correspond to that shown as broken line A in the figure and to be identical with the dimer of hexachlorocyclopentadiene prepared by reacting hexachlorocyclopentadiene with aluminum chloride as described in J. Am. Chem. Soc. 71, page 954, March 1949, in physical properties as well as its infrared spectrogram, shown as solid line B in the figure.

While the above describes the preferred embodiments of my invention, it will be understood that departures may be made therefrom within the scope of the specification and claims.

I claim:

1. A process for preparing dodecachlorotetrahydro-4,7-methanoindene which comprises heating decachlorotetrahydro-4,7-methanoindeneone with phosphorus pentachloride at temperatures between about 70° C. and about 160° C. for a period between about one hour and about six hours.

2. A process for preparing dodecachlorotetrahydro-4,7-methanoindene which comprises mixing decachlorotetrahydro-4,7-methanoindeneone with a quantity of PCl_5 at least the molecular equivalent of the decachlorotetrahydro-4,7-methanoindeneone and heating the resultant mixture to a temperature between about 70° C. and about 160° C. for a period of between about one hour and about six hours, cooling the resultant reaction mixture, drowning the mixture in water and recovering solid dodecachlorotetrahydro-4,7-methanoindene.

3. A process for preparing dodecachlorotetrahydro-4,7-methanoindene which comprises mixing substantially anhydrous decachlorotetrahydro-4,7-methanoindeneone with PCl_5 and heating the resulting mixture to a temperature between about 70° C. and about 160° C. for a period of between about one hour and about six hours, cooling the resultant reaction mixture, drowning the mixture in water, recovering solid dodecachlorotetrahydro-4,7-methanoindene and purifying the crude product by washing it with methanol, dissolving it in benzene and drowning the benzene solution in methanol to precipitate the purified product and recovering the crystalline dodecachlorotetrahydro-4,7-methanoindene.

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PLAINTIFF'S EXHIBIT 9

Nov. 26, 1957 E. E. GILBERT **Re. 24,397**

PROCESS FOR PREPARING DODECACHLOROTETRAHYDRO-4,7-METHANOINDENE

Original Filed April 6, 1951

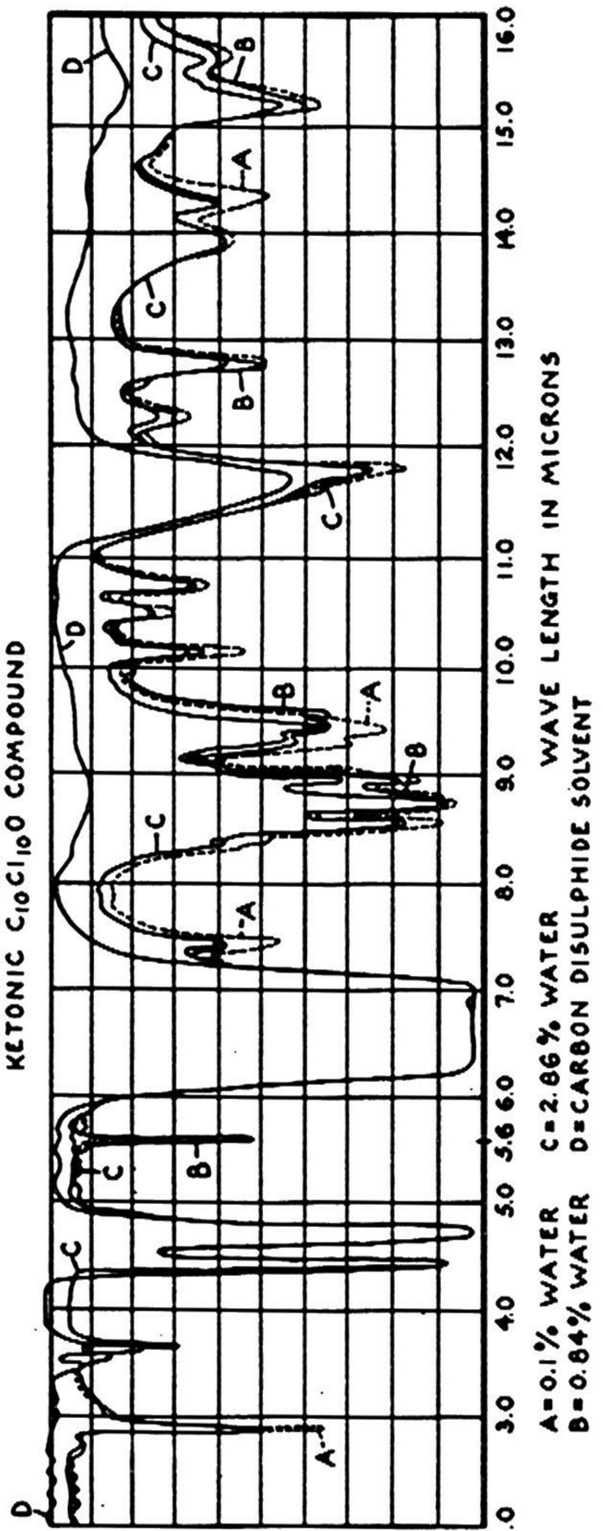

FIG.1.

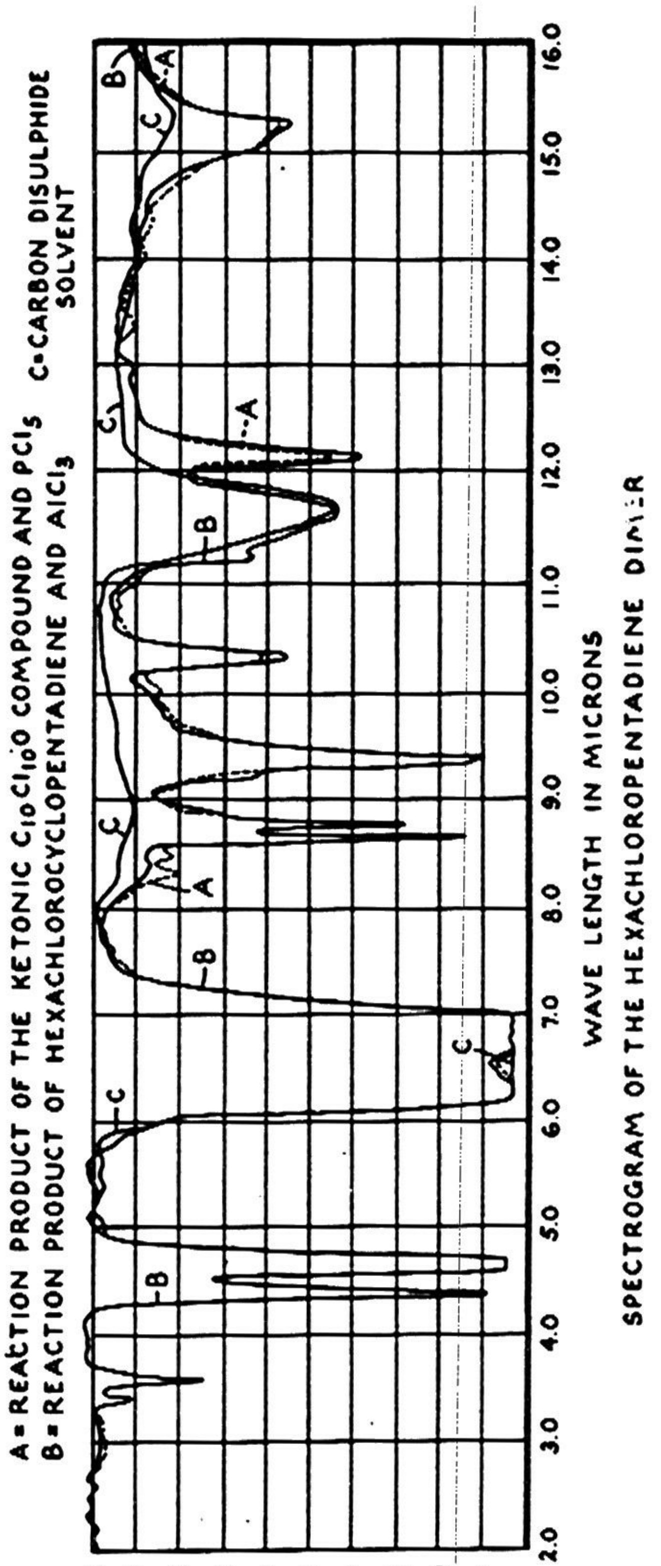

FIG.2.

INVENTOR
EVERETT E. GILBERT
BY
Elizabeth Hunter
ATTORNEY

UNITED STATES PATENT OFFICE

Certificate of Correction

Reissue No. 24,397 November 26, 1957

Everett E. Gilbert

It is hereby certified that error appears in the above numbered patent requiring correction and that the said Letters Patent should read as corrected below.

In the heading to the drawing sheet, line 2, and in the heading to the printed specification, lines 2 and 3, title of invention, for "PROCESS FOR PREPARING DODECACHLOROTETRAHYDRO-4,7-METHANOINDENE", each occurrence, read —PROCESS FOR PREPARING [DODECACHLOROTETRAHYDRO-4,7-METHANOINDENE] *A HEXACHLOROCYCLOPENTADIENE DIMER*—.

Signed and sealed this 15th day of April 1958.

[SEAL]

Attest:

KARL H. AXLINE,
Attesting Officer.

ROBERT C. WATSON,
Commissioner of Patents.

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United States Patent Office

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PROCESS FOR PREPARING DODECACHLORO-TETRAHYDRO-4,7-METHANOINDENE

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3 Claims. (Cl. 260—648)

Matter enclosed in heavy brackets [ ] appears in the original patent but forms no part of this reissue specification; matter printed in italics indicates the additions made by reissue.

This invention relates to a new method for preparing [dodecachhlorotetrahydro-4,7-methanoindene] a dimer of hexachlorocyclopentadiene by the reaction of phosphorus pentachloride with [decachlorotetrahydro-4,7-methanoindene-one] *the ketonic, hydrolyzed reaction product of hexachlorocyclopentadiene and sulfur trioxide, having the empirical formula $C_{10}Cl_{10}O$.*

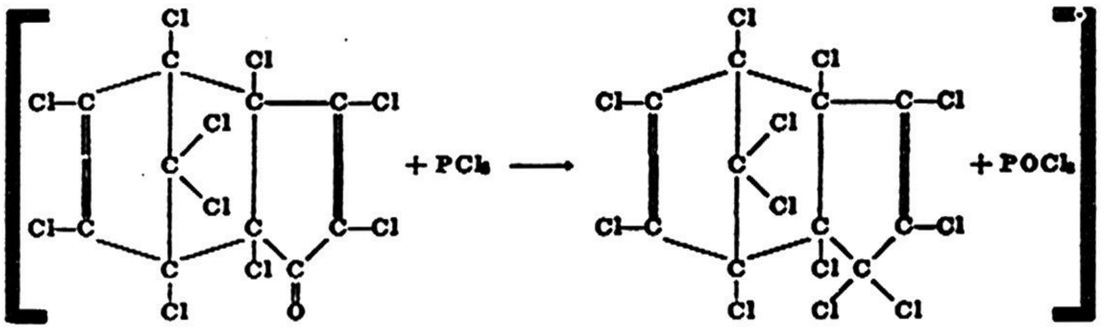
The hexachlorocyclopentadiene dimer *of my invention* has been prepared in the past by the condensation of hexachlorocyclopentadiene with aluminum chloride as described in J. Am. Chem. Soc. 71, page 954, March 1949, in the description of which process no structure was assigned to the hexachlorocyclopentadiene dimer.

I have now discovered the new process described below for preparing *this* hexachlorocyclopentadiene dimer [dodecachlorotetrahydro-4,7-methanoindene]. This *dimer* compound is useful as an intermediate in carrying out chemical reactions and as an insecticide.

The [decachlorotetrahydro-4,7-methanoindeneone] *ketonic $C_{10}Cl_{10}O$ compound* which forms the starting material for my new reaction may be prepared, for example, as described in my copending application with Silvio L. Giolito, Serial No. 196,123, filed November 17, 1950, now U. S. Patent 2,616,928 by condensing two molecules of hexachlorocyclopentadiene with the aid of sulfur trioxide to form a hexachlorocyclopentadiene-SO_3 reaction product and hydrolyzing the resulting reaction product to the ketone. It [is believed to be the 2,3,3a,4,5,6,7,7a,8,8-decachloro-3a,4,7,7a-tetrahydro-4,7-methanoindene-1-one illustrated below, and] usually exists in the form of a hydrate in a wide range of degrees of hydration.

The reaction as carried out according to my invention proceeds as follows:

$$C_{10}Cl_{10}O+PCl_5\rightarrow C_{10}Cl_{12}+POCl_3$$



In the drawings, [the single] *Figure 1 illustrates the infrared spectrogram of the ketonic $C_{10}Cl_{10}O$ compound which forms the starting material of my invention.* Figure 2 illustrates the infrared spectrogram of [dodecachlorotetrahydro-4,7-methanoindene] *the hexachlorocyclopentadiene dimer of my invention.* The dotted line A illustrates the spectrogram of the dimer as prepared according to the process of my invention; the solid line B illustrates the spectrogram of the dimer resulting as the reaction product of hexachlorocyclopentadiene with aluminum chloride. These spectrograms are substantially identical. Solid line C is the spectrogram of carbon disulfide used as solvent in preparing the spectrograms.

In carrying out the process of my invention, [decachlorotetrahydro-4,7-methanoindeneone] *the ketonic $C_{10}Cl_{10}O$ compound,* preferably in substantially anhydrous form or in as low a state of hydration as practicable, is reacted with phosphorus pentachloride to effect the replacement of the ketone group with chlorine, for example by mixing the reactants and heating the mixture. The resulting oily reaction product is then cooled and drowned in water, whereupon solid hexachlorocyclopentadiene dimer crystallizes and may be recovered as by filtration and, if desired, washed with a liquid such as methanol which is a solvent for *the* [decachlorotetrahydro-4,7-methanoindeneone] *ketonic $C_{10}Cl_{10}O$ compound,* but a non-solvent for the reaction product, to dissolve any unreacted [decachlorotetrahydro-4,7-methanoindeneone] *ketonic $C_{10}Cl_{10}O$ compound* which may be present. The crude dimer product may then be crystallized from a suitable solvent if desired, such as benzene, isopropanol or the like, and if desired may be further purified of residual [decachlorotetrahydro-4,7-methanoindeneone] *ketonic $C_{10}Cl_{10}O$ compound,* by first dissolving it in a hot hydrocarbon solvent such as benzene and then precipitating it out of solution by the addition of a hydrocarbon-miscible non-solvent for the [dodecachlorotetrahydro-4,7-methanoindene] *hexachlorocyclopentadiene dimer,* such as methanol, in which the [decachlorotetrahydro-4,7-methanoindeneone] *$C_{10}Cl_{10}O$ ketone* is soluble. The product may be further purified if desired, by recrystallization from an aliphatic alcohol such as isopropanol. The resulting [dodecachlorotetrahydro-4,7-methanoindene] *hexachlorocyclopentadiene dimer* is a white, crystalline solid which sublimes without melting at temperatures above about 240° C., is appreciably soluble in benzene, acetone, kerosene, carbon tetrachloride, etc. and while virtually insoluble in methanol, is appreciably soluble in the lower aliphatic alcohols having two or more carbon atoms, such as ethanol, isopropanol, etc., particularly at elevated temperatures.

The quantity of PCl_5 to be used in the preparation of the dimer is not critical, but for best yields should be at least the molecular equivalent of the [decachlorotetrahydro-4,7-methanoindeneone] *ketonic $C_{10}Cl_{10}O$ compound,* so as to furnish two chlorine atoms to replace the keto oxygen as indicated in the reaction above illustrated, and to react with any water of hydration present. Thus, although it is desirable to use [decachlorotetrahy-

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dro-4,7-methanoindeneone] *ketonic $C_{10}Cl_{10}O$ compound* starting material in as low a state of hydration as possible, nevertheless, hydrated forms may be used if a sufficient quantity of PCl_5 is employed to react with all the water of hydration as well as with the keto oxygen, forming $POCl_3$ and HCl, and when the expression "equimolecular quantities" is used in the claims to describe the proportions of reactants, it is to be understood to mean a quantity of PCl_5 sufficient to react with the keto oxygen and also with the water of hydration present. However, as the use of excesses of PCl_5 are not only wasteful, but entail handling of the troublesome HCl formed as a by-product of the reaction of PCl_5 with the water of hydration, it is preferable to dehydrate the [decachlorotetrahydro-4,7-methanoindeneone] *ketonic $C_{10}Cl_{10}O$ compound* as much as practicable before carrying out the reaction. In general, therefore, quantities of PCl_5 only slightly in excess of the molecular equivalent of the [decachlorotetrahydro-4,7-methanoindeneone] *ketonic $C_{10}Cl_{10}O$ compound* as defined are desirable.

The temperature to which the mixture of deca[chlorotetrahydro-4,7-methanoindeneone] *$C_{10}Cl_{10}O$ ketone* and PCl_5 is heated should be at least about 70° C., preferably between about 115° C. and about 160° C. The reaction is usually complete in a period of between about one hour and about six hours. The desired reaction product is separated from the resulting oily reaction mixture, for example by cooling and drowning in an excess of water, for example between about 5 and about 10 volumes of water per volume of the oily reaction mixture, whereupon solid [dodecachlorotetrahydro-4,7-methanoindene] *hexachlorocyclopentadiene dimer* crystallizes and may be separated by suitable means such as filtration, etc. The resulting crude product is purified by washing with a solvent for [decachlorotetrahydro-4,7-methanoindeneone] *the ketonic $C_{10}Cl_{10}O$ compound*, for example methanol.

The hexachlorocyclopentadiene dimer prepared according to my invention, has the infrared spectrogram shown as dotted line A in [the] Figure 2. It is identical with the spectrogram of the dimer of hexachlorocyclopentadiene prepared by reacting hexachlorocyclopentadiene with aluminum chloride as described in J. Am. Chem. Soc. 71, page 954, March 1949, as indicated by the infrared spectrogram of a material made by the $AlCl_3$ condensation method which is shown as solid line B in [the] Figure 2.

The infrared spectrograms shown in the figures were prepared on a standard infrared recording spectrophotometer designed for measuring and recording the infrared transmission of solids, liquids and gases, comprising a double infrared beam which scans the spectrum through the wave length range 2.0 to 16 microns, one part of the beam passing through the sample under study, the other passing through a compensating cell. If the sample under study absorbs radiation, the two beams become unequal. The magnitude of this inequality is a measure of the transmission of the sample of the particular wave length, and the record of these differences within the range of wave lengths scanned is the infrared spectrogram, recorded as an ink drawn line on a chart graduated in percent transmission as ordinates and in wave length as abscissae.

Solid samples, such as the compound of my invention, are conveniently measured in solution. The spectrogram shown in the figures was measured by dissolving 0.5 gram of the solid in carbon disulfide and diluting to 10 ml. with the solvent. A small amount of the solution was then introduced into a liquid cell with sodium chloride windows and sealed. The cell was placed in the spectrophotometer in the path of one of the beams as described above.

The infrared spectrogram of any chemical compound serves as an accurate means for identifying the compound. It has been compared with a human fingerprint in its ability to identify a compound with certainty. The characteristic reproducibility of the infrared spectrogram of a given compound is due to the facts that when a molecule is excited by infrared radiation it absorbs energy to a greater degree at some wavelengths than at others, and that the amount of absorption depends on the configuration and upon the linkages of the atoms composing the molecule. Accordingly, this spectrogram identifies and characterizes the hexachlorocyclopentadiene dimer with certainty.

The following specific example further illustrates my invention.

EXAMPLE I

Five parts of hydrated [decachlorotetrahydro-4,7-methanoindeneone] *$C_{10}Cl_{10}O$ ketone*, purified by solution in methanol, precipitation with water and drying, were mixed with 21 parts of phosphorus pentachloride and the mixture heated for three hours at 125°–150° C. The resulting reaction mixture was cooled to 20° C. and drowned in water. The solid material which formed was filtered and water washed, then washed with methanol to dissolve any unreacted [decachlorotetrahydromethanoindeneone] *$C_{10}Cl_{10}O$ ketone* which might be present, and dissolved in hot benzene. The benzene solution was mixed with a large excess of methanol to reprecipitate the product, which was filtered, dried and then recrystallized from isopropanol. Two parts of hexachlorocyclopentadiene dimer were obtained. An infrared spectrogram prepared from this product was found to correspond to that shown as broken line A in the figure and to be identical with the dimer of hexachlorocyclopentadiene prepared by reacting hexachlorocyclopentadiene with aluminum chloride as described in J. Am. Chem. Soc. 71, page 954, March 1949, in physical properties as well as its infrared spectrogram, shown as solid line B in the figure.

While the above describes the preferred embodiments of my invention, it will be understood that departures may be made therefrom within the scope of the specification and claims.

I claim:

1. A process for preparing [dodecachlorotetrahydro-4,7-methanoindene] *a hexachlorocyclopentadiene dimer having the infrared spectrogram shown in Figure 2 of the drawings* which comprises heating [decachlorotetrahydro-4,7-methanoindeneone] *the ketonic $C_{10}Cl_{10}O$ compound having the infrared spectrogram shown in Figure 1 of the drawings*, with phosphorus pentachloride at temperatures between about 70° C. and about 160° C. for a period between about one hour and about six hours.

2. A process for preparing [dodecachlorotetrahydro-4,7-methanoindene] *a hexachlorocyclopentadiene dimer having the infrared spectrogram shown in Figure 2 of the drawings* which comprises mixing *the ketonic $C_{10}Cl_{10}O$ compound having the infrared spectrogram shown in Figure 1 of the drawings*, [decachlorotetrahydro-4,7-methanoindeneone] with a quantity of PCl_5 at least the molecular equivalent of the *said ketonic compound* [decachlorotetrahydro-4,7-methanoindeneone] and heating the resultant mixture to a temperature between about 70° C. and about 160° C. for a period of between about one hour and about six hours, cooling the resultant reaction mixture, drowning the mixture in water and recovering solid [dodecachlorotetrahydro-4,7-methanoindene] *hexachlorocyclopentadiene dimer.*

3. A process for preparing [dodecachlorotetrahydro-4,7-methanoindene] *a hexachlorocyclopentadiene dimer having the infrared spectrogram shown in Figure 2 of the drawings* which comprises mixing *the* substantially anhydrous [decachlorotetrahydro-4,7-methanoindeneone] *ketonic $C_{10}Cl_{10}O$ compound having the infrared spectrogram shown in Figure 1 of the drawings*, with PCl_5 and heating the resulting mixture to a temperature between about 70° C. and about 160° C. for a period of between about one hour and about six hours, cooling the resultant

reaction mixture, drowning the mixture in water, recovering solid [dodecachlorotetrahydro-4,7-methanoindene] *hexachlorocyclopentadiene dimer* and purifying the crude product by washing it with methanol, dissolving it in benzene and drowning the benzene solution in methanol to precipitate the purified product and recovering the crystalline [dodecachlorotetrahydro-4,7-methanoindene] *hexachlorocyclopentadiene dimer*.

References Cited in the file of this patent or the original patent

Hunter et al.: "Jour. Am. Chem. Soc.," vol. 55, pages 2567–70 (1933).

Degering et al.: "An Outline of Organic Chemistry," third edition, pages 56–7, 200 (1939).

"Chemical Abstracts," vol. 35, column 2469 (1941). Abstract of article by Churbakov.

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A. F. Holleman--LL. D., D. Sc., F. R. S. E.
ORGANIC CHEMISTRY
(By J. P. Wibaut)

TABLE 5

PARTITION COEFFICIENTS

	Partition coefficient between water and				
	ether	petroleum ether	benzene	chloroform	methyl ethyl ketone
Acetic acid	2.1 (25°)	200	28 (25°)	15 (25°)	0.9 (25°)
n-Butyric acid	0.16 (25°)	9	0.18	0.8 (25°)	0.03 (25°)
n-Valeric acid	0.04 (22°)	10.2			0.1 (25°)
n-Caproic acid	0.01 (23°)	0.2	0.08	0.03 (25°)	
Oxalic acid	9 (15°)				
Succinic acid	6 (15°)				
Lactic acid	11 (27°)				
Tartaric acid	250 (27°)				
Benzoic acid	0.014	1.0 (20°)		0.08	
Salicylic acid	0.004 (15°)		0.4 (25°)	0.3 (25°)	
Phenol			0.5 (25°)	0.3 (25°)	
Picric acid	0.5	> 1	0.5 (20°)	0.3 (25°)	
m-Nitrobenzoic acid	0.1		0.4 (20°)	0.06	
Methylamine	> 1		25 (25°)	12	
Trimethylamine	2.4 (20°)		2.0		

Separation by fractional crystallisation is impossible, if the components of the mixture form mixed crystals, since in this case the crystals which separate are composed of both components.

(*ii*) CHARACTERISTICS OF PURITY AND THE IDENTIFICATION OF ORGANIC COMPOUNDS BY MEANS OF PHYSICAL CONSTANTS

10. In principle the determination of any physical constant can serve as a characteristic for the purity of a chemical compound. The physical constants which come into consideration for this purpose are, for example, the melting point, the boiling point, with liquids the specific gravity and the refractive index, the optical rotatory power (of optically active substances). Determination of the crystalline form may also be of considerable help in identifying a substance.

The *determination of the melting point* has great practical significance. A mixture of two (or more) substances does not melt at one temperature, but the melting takes place over a temperature range; the temperature at which everything is melted, usually lies lower than the melting temperature of either of the substances separately. An exception to this rule can occur only with substances which form a continuous series of mixed crystals. Hence, if a substance is recrystallised—preferably from different solvents—

until the melting point no longer changes on repeating the process, it may be concluded, with a high degree of probability, that the preparation consists of one chemical compound. Further, it follows, that if a mixture of two substances A and B shows the same melting point as each of the substances A and B separately, these substances are identical.

On this fact depends the ***mixed melting point test***, which is very frequently used to decide whether two crystalline preparations, which are expected to consist of the same chemical compound, are actually indentical.

In observing melting points and drawing conclusions from them, account has to be taken also of the possibility of *polymorphism*, by which is understood, that the same chemical compound can occur in different crystalline modifications, showing different melting points. A number of examples of such dimorphous substances are already known among organic compounds.

In the practice of organic-chemical research it is in many cases sufficient to determine the melting point with an accuracy of 0.5 to 1.0° C. 10–20 Milligrams of the substance are introduced into a narrow, thin-walled, glass tube, sealed at one end, which is then fixed to a thermometer by means of a small rubber ring, in such a way that the substance is situated at the same height as the mercury reservoir. The thermometer is then placed in a high-boiling liquid (paraffin oil); the melting point apparatus is set up in such a way that the temperature of the liquid surrounding the thermometer slowly and regularly rises. The temperature is read off at the moment when the crystals melt. Sometimes, it is preferable to determine the setting point instead of the melting point. The test is then carried out with a few grams of substance; the thermometer is placed in the molten substance.

The determination of the boiling point is usually carried out in a fractionating flask, from which the liquid is at the same time distilled. It also applies here, that a mixture does not boil at one definite temperature, but within a range of temperatures. Now, however, the temperature at which the mixture begins to boil, may be either higher or lower than the boiling point of the pure substance, according to whether the impurity boils higher or lower; this is clearly apparent from a consideration of the boiling-points curve (see p. 24). A boiling point is not so easily determined with an accuracy of 0.1° as a melting point or a setting point, partly on account of the effect of atmospheric pressure. With liquids the determination of the specific gravity and of the refractive index can be a very good criterion of the purity.

11. *Specific gravity.* The specific gravity of organic liquids alters considerably with the temperature, since the coefficient of expansion of organic liquids is nearly always much greater than that of water. The specific gravity is usually measured with a pycnometer, (cf. Fig. 12).

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DEFENDANT'S EXHIBIT 2

A TEXT-BOOK OF

PRACTICAL ORGANIC CHEMISTRY

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QUALITATIVE ORGANIC ANALYSIS

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By

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With diagrams and 8 photographs

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temperature at which the solution just becomes homogeneous is taken as the critical solution temperature. The same result is obtained by observing the temperature at which the second phase appears. The water content may then be deduced with the aid of the following table, and checked, if desired, by a determination of the density (Section **XI,2**).

PURITY OF CH_3OH (%)	C.S.T.	$d_4^{20°}$
100·0	36·55°	0·79578
99·80	37·90	0·79634
99·65	38·95	0·79676
99·50	40·05	0·79718
99·35	41·00	0·79760
99·05	43·00	0·79845
98·69	45·00	0·79947
98·41	46·25	0·80026
98·01	48·05	0·80140
97·48	50·15	0·80292
97·13	51·10	0·80390

For ethyl alcohol, two volumes of di*cyclo*hexyl * are mixed with one volume of the alcohol, a thermometer is introduced, and the mixture heated until it becomes clear. The solution is then slowly cooled, with constant stirring, and the temperature is determined at which the opalescent solution suddenly becomes turbid so that the immersed portion of the mercury thread of the thermometer is no longer clearly visible. This is the C.S.T. The water content may then be evaluated by reference to the following table.

PURITY OF C_2H_5OH (%)	C.S.T.	$d_4^{20°}$
100·0	23·4°	0·78934
99·9	25·4	0·78966
99·8	27·3	0·78997
99·7	29·2	0·79028
99·6	31·0	0·79059
99·5	32·8	0·79089
99·0	41·0	0·79243
98·5	48·0	0·79346

THEORY OF MELTING AND FREEZING

I,10. Melting point and vapour pressure. The melting point of a crystalline solid is the temperature at which the solid begins to change into liquid under a pressure of one atmosphere. For pure substances, the change from the solid to the liquid state is quite sharp (within 0·5°), hence the temperature is valuable for purposes of identification. Moreover, the melting point is considerably influenced by the presence of other

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* This may be prepared by the catalytic reduction of pure diphenyl (see Section III, 150.) The Eastman Kodak product is satisfactory.

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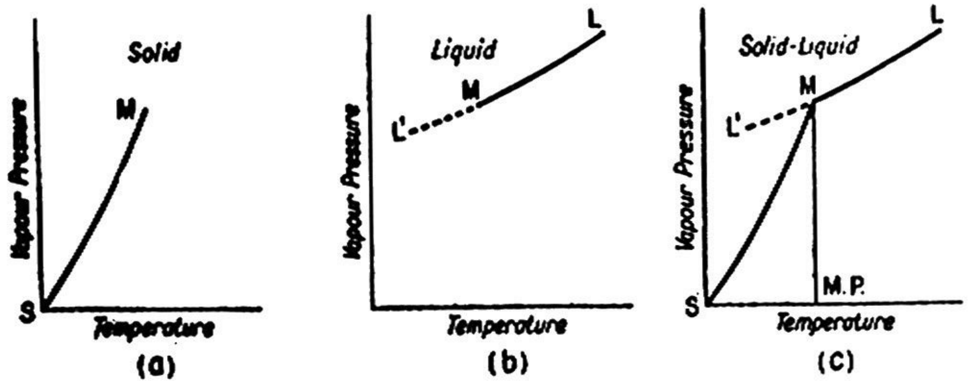
substances, and it is therefore an important criterion of purity. If the liquid is cooled, solidification will occur at the same temperature,* and for a pure substance the melting point and freezing point are identical. The freezing point is usually defined as the temperature at which both liquid and solid can exist in contact with each other under a total pressure of one atmosphere.

The reason for the constancy and sharpness of the melting point of a pure crystalline solid can be appreciated upon reference to Fig. *I*, *10*, 1, in which (*a*) is the vapour pressure curve of the solid and (*b*) that of the liquid form of the substance. Let us imagine a vessel, maintained at constant temperature, *completely* filled with a mixture of the above liquid and solid. The molecules of the solid can only pass into the liquid and the molecules of the liquid only into the solid. We may visualise two competitive processes taking place: (i) the solid attempting to "evaporate" but it can only pass into the liquid, and (ii) the liquid attempting to "distil" but it can only pass into the solid. If process (i) is faster, the solid will melt, whereas if process (ii) proceeds with greater speed the substance would completely solidify. To decide which process is likely to occur at a particular temperature, curve (*a*) must be superimposed upon curve (*b*) thus giving curve (*c*). The rate of rise of vapour pressure with temperature is greater for the solid than for the liquid, consequently the two curves will intersect. At the intersection (and only at this temperature), both solid and liquid can coexist; this is therefore the freezing point or melting point since these two constants are identical for a pure substance. Provided the pressure is atmospheric, the temperature *M* is an absolute constant. If the temperature is less than *M*, the liquid will pass more rapidly into solid than the solid into liquid and hence the substance will remain solid, whilst for temperatures greater than *M* the solid, having the greater vapour pressure, will pass more rapidly into the liquid phase. It is only at the temperature *M*, at which the vapour pressures of the liquid and the solid are equal in magnitude, that the rates of transformation are identical and both liquid and solid can coexist, provided the temperature is maintained constant. As soon as the



(a) (b) (c)

Fig. *I*, *10*, 1.

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* A liquid can sometimes be supercooled, *i.e.*, the temperature can be reduced below the freezing point without solid separating, but as soon as solid does appear the temperature immediately rises to the true freezing point.

temperature is raised even a fraction of a degree above *M*, the solid will be completely converted into the liquid phase, assuming of course that sufficient time is allowed for the transformation to take place. This is why the rate of rise of temperature in the vicinity of the melting point must be small (say, 1–2° per minute) for an accurate determination of this constant.

It is a well known fact that substances like water and acetic acid can be cooled below the freezing point: in this condition they are said to be **supercooled** (compare supersaturated solution). Such supercooled substances have vapour pressures which change in a normal manner with temperature: the vapour pressure curve is represented by the dotted line *ML'*—a continuation of *ML*. The curve *ML'* lies above the vapour pressure curve of the solid and it is apparent that the vapour pressure of the supersaturated liquid is greater than that of the solid. The supercooled liquid is in a condition of metastability. As soon as crystallisation sets in, the temperature rises to the true freezing or melting point. It will be observed that no dotted continuation of the vapour pressure curve of the solid is shown; this would mean a suspended transformation in the change from the solid to the liquid state. Such a change has not been observed nor is it theoretically possible.

I,11. Effect of impurities upon the melting point. Let us take a specific example and examine the effect of the addition of a small quantity of naphthalene to an equilibrium mixture of pure solid and liquid α-naphthol at a temperature of the true melting point (95·5°) at atmospheric pressure. The naphthalene will dissolve in the liquid α-naphthol and, according to Raoult's law, the vapour pressure of the latter will be reduced. Hence α-naphthol will pass preferentially into the liquid phase and, if the temperature is maintained at 95·5°, the ultimate result will be the complete melting of the solid α-naphthol; since melting requires heat and no heat is imparted to the system, the temperature will fall. The effect can easily be interpreted by reference to Fig. *I*, *11*, 1. The initial state of the system is represented by *M*. Upon the addition of a small amount of the second substance, the vapour pressure of the *liquid* first falls to *A*. Now a solid phase with vapour pressure *M* cannot coexist with a liquid phase with lower vapour pressure *A*. The solid represented by *M* will therefore melt and tend to pass into the condition expressed by *A*. However, in melting heat is absorbed, and the temperature will consequently fall. Thus the vapour pressures of the solid *M* and the liquid *A* will fall along their respective vapour pressure curves, which will intersect at M_1. At M_1 equilibrium results and no further change will take place; here solid α-naphthol and the solution of naphthalene in α-naphthol may coexist indefinitely—this, it will be observed, is below the melting point of the pure substance (α-naphthol). Further addition of naphthalene

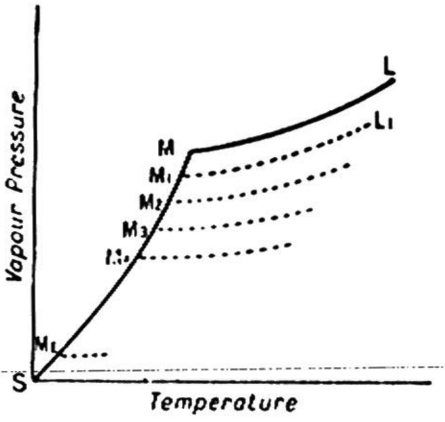

Fig. *I*, *11*, 1.

DEFENDANT'S EXHIBIT 2

DEFENDANT'S EXHIBIT 3

LABORATORY TECHNIQUE IN ORGANIC CHEMISTRY
(By Avery Adrian Morton)

on the walls of the unwashed tubes as low as 150°, whereas in cleaned tubes the melting point was sharp at 165°.

30. The Rate of Heating.—Time must be allowed for transfer of heat through the glass wall of the capillary tube to the organic solid and for dissipation of the heat of fusion. This time is reduced to a minimum by using thin-walled tubing. Because observations of thermometer and sample are not simultaneous, the rate of heating must be slow enough to make the error from this source negligible. An interesting effect from too rapid heating has been recorded by Letsche[5] who observed that the melting point of glycocholic acid could be changed from 132 to 178° by varying the rate of heating. A rise of 1° a minute at the melting point is about the maximum rate at which it is possible to make satisfactory observations of the change of state. In the apparatus of Francis and Collins[4] for very exact determinations (see Sec. 38) the rate during fusion was 0.1° in 4 min.

31. Relation of the Purity to Melting Point.—The *sharpness* with which a compound melts is assumed to be an index of its purity. Fortunately this assumption is usually correct when apparatus and manipulation are satisfactory. A rare exception occurs when the material under investigation is a eutectic mixture. Trouble is sometimes experienced with the presence of a constant amount of impurity which causes the temperature to fall a trifle short of that usually attainable. By recrystallizing from another solvent this error may frequently be avoided. Indeed, as a general precaution the observation of the melting point of a material crystallized from two different solvents can be highly recommended as a check on purity. If this method fails to produce a sharp melting point, the product should be distilled, sublimed, steam distilled, extracted, treated with decolorizing carbon, chromatographically adsorbed, or otherwise handled in a further attempt to eliminate the trace of impurity. Occasionally, mixtures melt as sharply as a single material. A molecular compound which resisted attempts at separation by recrystallization from different solvents was reported by Kohler and Mydans.[7] Oxotetraphenylpropanol (m. p. 150°) with another mol of tetraphenylpropenone (m.p. 154°) formed an addition compound melting sharply at 162°. It was separated by subjecting it to a chemical reaction which differentiated between the components.

While the sample with *highest melting point* would obviously represent highest purity for any given material, it does not follow that the highest melting point given in the chemical literature represents the purest specimen. Tseng[6] reports discrepancies that amounted to more than 2° with different types of apparatus and differences of as much as 1° with different observers using the same apparatus. These figures represent probably the maximum amount of divergence. Francis and Collins[4] state that they seldom differed from each other by more than 0.2° using an ordinary apparatus, a bath of sulfuric acid heated by means of a Bunsen flame, and a thermometer on which 1 mm. of scale length was equivalent to 1°. With a better apparatus (see Sec. 38), carefully prescribed conditions, and a thermometer with a more open scale, they found a difference among observers no greater than 0.03°. This value probably represents the maximum amount of reproducibility so far attained by this method. It is clear that values recorded in the literature are not always accurate and that the specimen reported to have the highest melting point need not necessarily be the purest unless accompanied by full description of precautions observed.

32. Mixed Melting Points.—Another common assumption is that mixtures of unlike substances will melt lower than the individual members. So general is this belief that failure of a mixture to show a depressed melting point is taken as a criterion of the identity of two substances. Again, it is fortunate that the rule is of such wide applicability. Such a generality should not obscure the fact that systems do occur in which no change or an elevation of melting point is realized. This statement is merely equivalent to saying that the melting points of all mixtures of organic compounds cannot be represented by the diagram shown in Fig. 4*a* in which the substances *A* and *B* crystallize from solution substantially uncontaminated with the other until the eutectic composition is reached. Systems of mixed crystals (Fig. 4*b*) or of those in which compound formation occurs (Fig. 4*c*) are equally possible. That the rule of mixed melting points

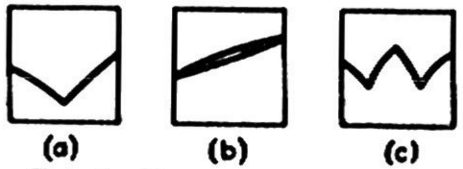

Fig. 4.—Temperature-composition diagrams showing typical behavior upon solidification.

DEFENDANT'S EXHIBIT 4

NATURAL AND SYNTHETIC HIGH POLYMERS
A Textbook and Reference Book For Chemists and Biologists
(By Kurt H. Meyer, Ph.D)

given by Waentig and Peschek.[21] The solubility of palmitic acid in carbon tetrachloride at 0° C. is 0.5%, while in a solution of 1% lauric acid in the same solvent, palmitic acid is twice as soluble.

In the case of truly homogeneous, well-crystallized high polymers, such as frequently occur in nature, solubility is independent of the quantity of solid phase present. The constancy of solubility and the independence of solubility of the quantity of solid phase present are useful criteria for testing the purity of proteins (Cohn,[22] Northrop[22]). This is analogous to using the constancy of melting point as a test for purity in compounds of low molecular weight.

Many commercial preparations of high polymers consist of mixtures of substances differing in solubility, and are therefore partially soluble in certain solvents. In contrast to normal usage, the term "solubility" is frequently used to describe the proportion of soluble substance present and is expressed as a percentage of the material. Thus "10% alcohol-soluble nitrocellulose" describes a sample containing 10% of alcohol-soluble constituents.

3. The Influence of Individual Groups on Solubility

No quantitative data for the action of individual groups on particular solvents and hence for the behavior of high polymers composed of such groups are available. Nevertheless, very promising beginnings have been made by Hildebrand,[23] by Wolf and his collaborators, and by Briegleb in the systematic study of intermolecular forces and their influence on the state of solution and on solubility in compounds of low molecular weight; these investigations are described in detail in the preceding volume of this series. Roughly speaking, the various groups may be divided into hydrophilic groups (which are always polar) and hydrophobic groups; the hydrophilic groups are lipophobic, that is, are insoluble in apolar, "lipoid" solvents, while the hydrophobic groups are lipophilic. The ratio of hydrophilic to hydrophobic groups in a molecule determines its solubility (and the swelling powers of the substance) in water, on the one hand, and in organic solvents, on the other, since the heat of solution will be derived, approximately additively, from the increments due to the individual groups. (Geometrical or steric factors may be of importance in hindering the reaction of certain groups with the solvent.)

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[21] P. Waentig and G. Peschek, *Z. physik. Chem.*, 93, 529 (1919).

[22] See footnotes 28, page 432, and 29, page 433.

[23] J. H. Hildebrand, *Solubility of Non-electrolytes.* Reinhold, New York, 1936.

It is therefore understandable that polymeric hydrocarbons dissolve in hydrocarbons; high-polymeric olefines dissolve particularly readily, with evolution of heat, in halogenated hydrocarbons such as $CHCl_3$, which are also miscible with olefines of low molecular weight. Substances with many hydrophilic groups, such as polyacrylic acid, proteins, and polyvinyl alcohol, are soluble in water, formamide and similar solvents; on the other hand, substances which, like the esters of cellulose, contain hydrophobic residues along with hydrophilic groups, dissolve in solvents or mixtures also containing both hydrophilic and lipophilic groups. These general points have been emphasized by Whitby,[24] Lee,[25] and Sheppard and Sweet,[26] and a large body of experimental evidence has been collected. On the other hand, the search for a relationship between solvent powers and dipole moment of the solvent appears to be hopeless, since both substances with apolar, hydrophobic groups and those with hydrophilic groups may, in spite of profoundly different solvent powers, possess the same dipole moment. Dioxan, in which the dipole moments of the two polar ether groups exactly compensate each other, offers an example of a solvent markedly hydrophilic in character but with zero dipole moment.

4. Gelation[27]

Solutions of many polymers and of many substances of low molecular weight (such as soaps) set, under certain conditions, to form translucent gels. In spite of a high liquid content, these have considerable mechanical rigidity; on the other hand, the mobility of the ions dissolved in the gel is inappreciably affected, as is evident from conductivity measurements. This leads one to conclude that such gels consist of a wide-meshed, solid structure, in the meshes of which ions and fluid particles are free to move.

Reversible and irreversible gel-formation can be distinguished by the ability or non-ability of the gel to return to the liquid condition.

(a) *Irreversible Gelation*

Typical irreversible gelation is exhibited by silicic acid. When liberated from solutions of alkali silicates, the acid polymerizes - as is shown by the

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[24] G. S. Whitby, *Colloid Symposium Monograph*, 4, 203 (1926).

[25] W. B. Lee, *J. Soc. Chem. Ind. Japan*, 49, 226 T (1931).

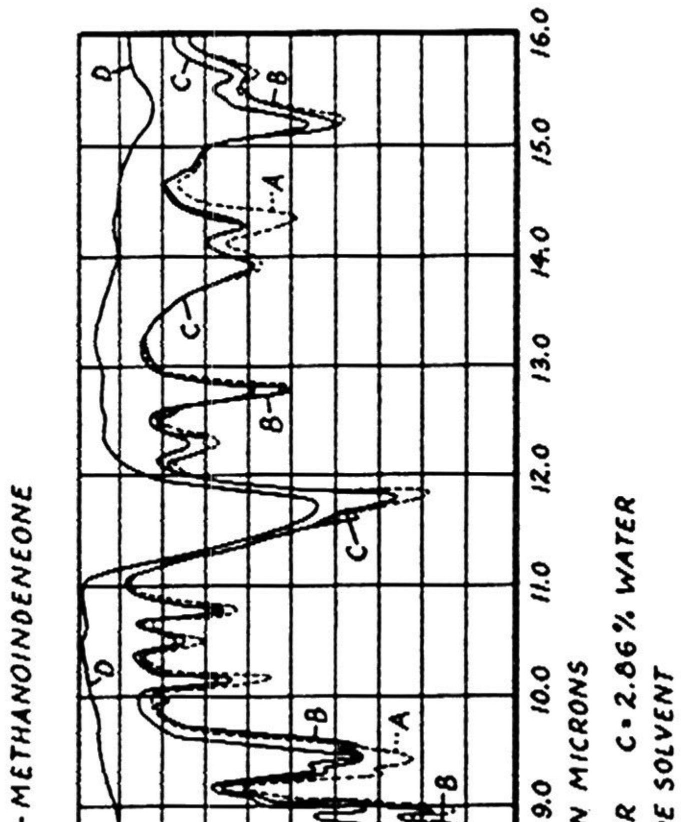
[26] S. E. Sheppard and S. S. Sweet, *J. Phys. Chem.*, 36, 819 (1932).

[27] Review: E. Heymann, *The Sol-Gel Transformation.* Hermann, Paris, 1936.

DEFENDANT'S EXHIBIT 5

Nov. 4, 1952 E. E. GILBERT ET AL **2,616,825**

DECACHLOROTETRAHYDRO-4,7-METHANOINDENEONE PESTICIDE AND
METHOD FOR COMBATING NOXIOUS ORGANISMS THEREWITH
Filed Nov. 17, 1950



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DEFENDANT'S EXHIBIT 5

Patented Nov. 4, 1952 **2,616,825**

UNITED STATES PATENT OFFICE

2,616,825

DECACHLOROTETRAHYDRO-4,7- METHANOINDENEONE PESTICIDE AND METHOD FOR COMBATING NOXIOUS ORGANISMS THEREWITH

Everett E. Gilbert, Flushing, and Silvio L. Giolito, New York, N. Y., assignors to Allied Chemical & Dye Corporation, New York, N. Y., a corporation of New York

Application November 17, 1950, Serial No. 196,121

11 Claims. (Cl. 167—30)

1

This invention relates to a new pesticidal composition comprising decachlorotetrahydro-4,7-methanoindeneone and to a method for combating noxious organisms therewith.

The decachlorotetrahydro-4,7-methanoindeneone compositions of our invention are useful in combating a wide variety of noxious organisms including insects and their larvae of the Hymenoptera (flies), Lepidoptera (moths), Coleoptera (beetles) and Orthoptera (grasshoppers) species, and also in combating crop-attacking fungi.

The compositions of our invention comprise essentially a decachlorotetrahydro-4,7-methanoindeneone, usually in hydrate form, which may be applied alone or in association with a spray or dust carrier material in any suitable manner adapted to combat the particular organism to be controlled. As a spray or impregnant, the solid decachlorotetrahydro-4,7-methanoindeneone may conveniently be dissolved or dispersed in any standard liquid carrier, such as kerosene or the like. As a dust, it may be mixed with a suitable finely divided solid material, such as clay or the like. It may readily be applied from solution in solvents in which it is soluble as more fully set forth hereinafter, for example in acetone, lower aliphatic alcohols, or in mixtures of such alcohols with water or kerosene. Application of decachlorotetrahydro-4,7-methanoindeneone in liquid solution form is especially adayted to moth-proofing of fabrics and to control of houseflies and the like, while application in the form of dusts or liquid sprays may conveniently be used in its application to vegetable crops and the like, to control infestations of insects or fungi, whereas grain or flour infesting organisms may be controlled by the addition of small quantities of undiluted decachlorotetrahydro-4,7methanoindeneone directly to the grain material.

The decachlorotetrahydro-4,7-methanoindeneone hydrate is characterized by the following chemical analysis:

	Percent Theory ($C_{10}Cl_{10}O$)	Percent Found
Cl	72.3	72.7
C	24.4	23.8
H	0	0.6
H_2O	0	0.1
Mol Wt	491	296

2

The decachlorotetrahydro-4,7-methanoindeneone is a white crystalline solid with no appreciable odor. Upon heating in a glass melting point tube by conventional procedures up to 300° C., no tendency to melt is noted. It sublimes when heated in the atmosphere; for example, slight sublimation occurs upon oven drying at 110°-115° C., while upon heating at 140° C. at 1-1.5 mm. of Hg pressure, 10-15% of its weight sublimes in three hours. It sublimes with some decomposition when heated in the open atmosphere to 300° C. It is readily soluble in acetone, lower aliphatic alcohols, ethers and the like, and also in nitrobenzene, and sulfuryl chloride. It is somewhat less soluble in benzene, toluene, hexane and petroleum ether, but is sufficiently soluble in warm hexane to allow the use of this material as a recrystallizing solvent if desired. It is virtually insoluble in cold water and only slightly soluble (less than 0.4%) in boiling water. It tends to gel upon separation by cooling, from hot solutions in hydrocarbon solvents. A satisfactory recrystallizing solvent is 85%-90% aqueous ethanol from which gelatin does not occur. It usually exists as a crystalline hydrate when exposed to atmospheric conditions and is useful for insecticidal, etc., purposes in hydrate form.

The decachlorotetrahydro-4,7-methanoindeneone compound is soluble in, and relatively stable toward, strong caustic solutions, such as sodium, potassium and calcium hydroxides. This stability towards caustic materials is of advantage in its application as a pesticide in combination with lime and other alkaline agricultural chemicals, and in this respect it is superior to benzene hexachloride (BHC) and dichlorodiphenyltrichloroethane (DDT) which decompose readily on contact with alkalis. Furthermore, being volatile under normally atmospheric conditions only at temperatures considerably above those usually encountered in use, it has a high residual insecticidal activity and because of its limited solubility in hydrocarbon solvents, it is considerably more resistant in dry cleaning than DDT, which is designated above in the use of this compound as a moth-proofing agent.

In the accompanying figure, the several lines represent the infared spectrogram of the decachlorotetrahydro-4,7-methanoindeneone, which is the essential toxic ingredient of the new pesticidal composition of our invention. The spectrograms were obtained in carbon disulfide solutions

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line A being the record of substantially anhydrous material (0.10% or 0.03 mol H_2O); solid line B being the record of a slightly hydrated sample (0.84% or 0.24 mol H_2O); while solid line C is the record of an essentially monohydrated sample (2.86% or 0.8 mol H_2O). The three spectrograms are considered substantially identical. Solid line D is the spectrogram of the CS_2 solvent.

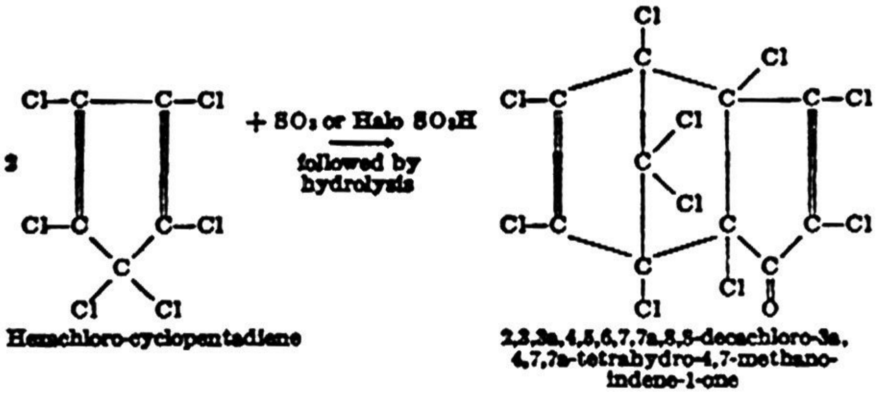
These infrared spectrograms were prepared on a standard infrared recording spectrophotometer designed for measuring and recording the infrared transmission of solids, liquids and gases, comprising a double infrared beam which scans the spectrum through the wave length range 2.0 to 16 microns, one part of the beam passing through the sample under study, the other passing through a compensating cell. If the sample under study absorbs radiation, the two beams become unequal. The magnitude of this inequality is a measure of the transmission of the sample of the particular wave length, and the record of these differences within the range of wave lengths scanned is the infrared spectrogram, recorded as an ink drawn line on a chart graduated in percent transmission as ordinates and in wave length as abscissae.

Solid samples, such as the compound of our invention and the related compounds described, are conveniently measured in solution. The spectrograms shown in the figures were all measured by dissolving 0.5 gram of the solid in carbon disulfide and diluting to 10 ml. with the solvent. A small amount of the solution was then introduced into a liquid cell with sodium chloride windows and sealed. The cell was placed in the spectrophotometer in the path of one of the beams as described above.

The infrared spectrogram of any chemical compound serves as an accurate means for identifying the compound. It has been compared with a human fingerprint in its ability to identify a compound with certainty. The characteristic reproducibility of the infrared spectrogram of a given compound is due to the facts that when a molecule is excited by infrared radiation it absorbs energy to a greater degree at some wave length than at others, and that the amount of absorption depends on the configuration and upon the linkages of the atoms composing the molecule. Accordingly, the compound is identified and characterized with certainty by its infrared spectrogram.

The new pesticidal composition of our invention may be prepared by any suitable process, for example by condensing two molecules of hexachlorocyclopentadiene with the aid of sulfur trioxide to form a hexachlorocyclopentadiene reaction product and hydrolyzing the reaction product to the ketone as described in copending application Serial No. 196,123 filed November 17, 1950; or by reacting hexachlorocyclopentadiene with a halosulfonic acid, such as chlorosulfonic acid and fluosulfonic acid, and hydrolyzing the resulting reaction product to form the ketone.

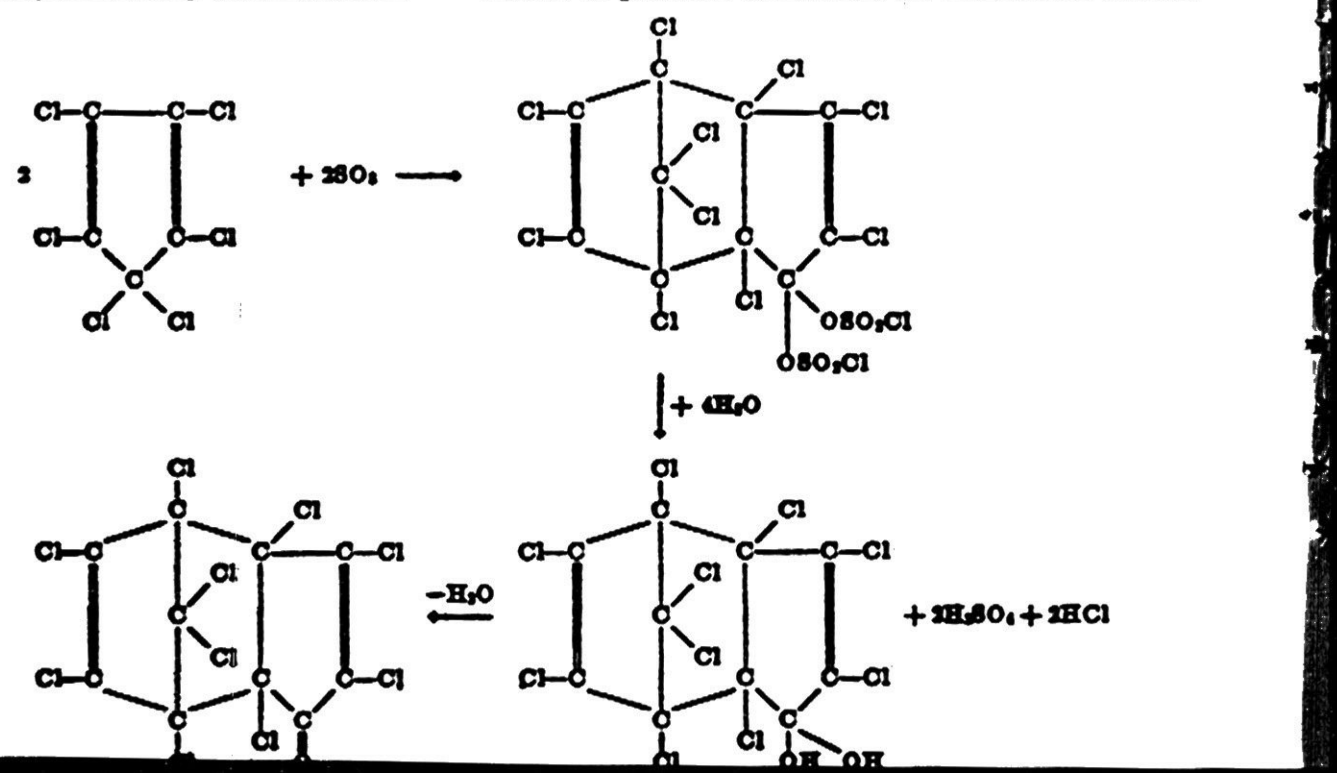
The exact mechanism of the reactions involved in the preparation of the decachlorotetrahydro-4,7-methanoindeneone product is not clearly understood, but the overall reaction is indicated in the equation set forth below:



Hexachloro-cyclopentadiene

2,3,3a,4,5,6,7,7a,8,8-decachloro-3a,4,7,7a-tetrahydro-4,7-methanoindene-1-one

While the overall reaction in the methods of preparation described above proceeds from the same starting material to the same end product when carried out using the SO_3 treatment and when using the halosulfonic acid treatment, the reaction mechanisms probably differ and are believed to proceed somewhat as illustrated below:



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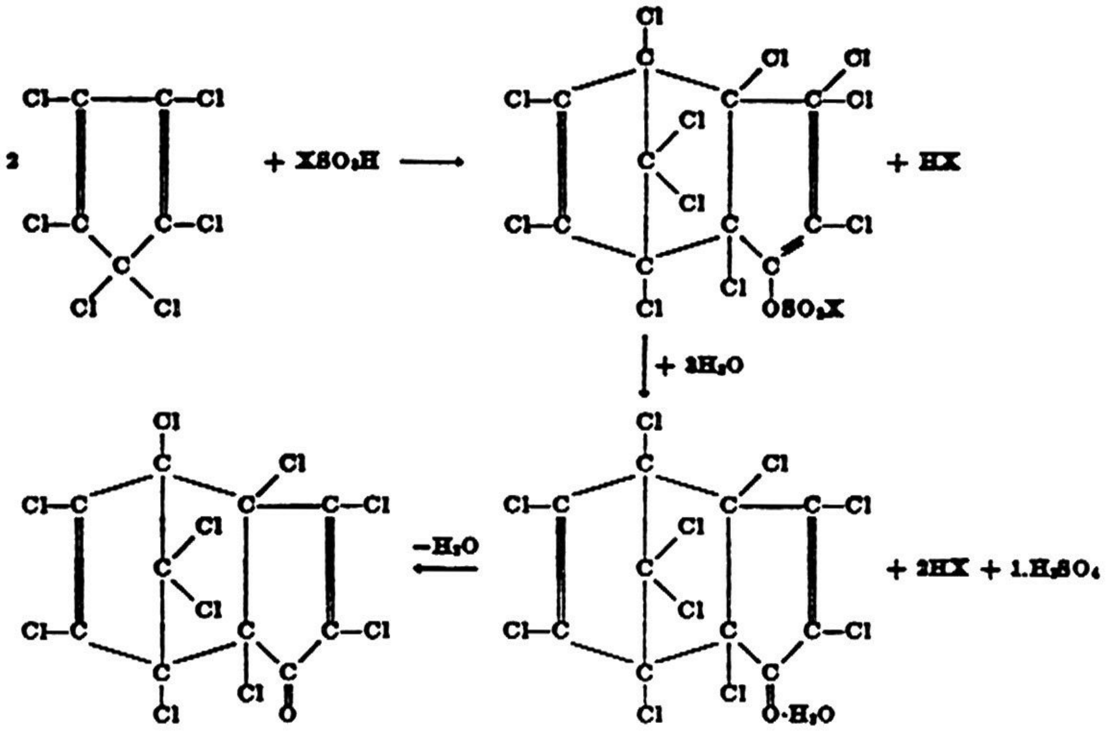

wherein X=chlorine or fluorine

In carrying out the process for preparing the decachlorotetrahydro - 4,7 - methanoindeneone compound above described, according to the procedure wherein sulfur trioxide is used, hexachlorocyclopentadiene and sulfur trioxide are mixed by charging them, either simultaneously or separately, to a reaction vessel. After mixing the reactants, the charge may be digested for a short period, sufficient to condense two molecules of hexachlorocyclopentadiene and to form a liquid reaction product of hexachlorocyclopentadiene and sulfur trioxide.

The liquid SO_3-hexachlorocyclopentadiene reaction product thus formed is then hydrolyzed to the decachloromethanoindeneone, by drowning the reaction product in a relatively large volume of an aqueous drowning medium, preferably an alkaline aqueous medium, for example in at least about 5 volumes of drowning medium per volume of reaction product.

After addition of the reaction product to the drowning medium is complete, the mixture may be digested for a short additional period, for example a half hour to insure completion of the hydrolysis.

The product may be filtered at this stage, if desired, but to improve coagulation and ease of filtration, we prefer to agitate the batch for a short period at a temperature somewhat higher than the drowning temperature, for example at about 90°–95° C. for about a half hour, during which the crystalline product may partially or completely redissolve.

When the drowning has been effected in an alkaline solution, the charge is then neutralized, for example with sulfuric acid, whereupon the crystalline product reprecipitates. The amount of acid added for neutralization should be sufficient to bring the mixture preferably to neutrality, or to a very slight alkalinity not exceeding about 0.05% NaOH, as indicated by a pH of 7–8.

After neutralization, the charge is agitated, preferably at a temperature between about 90° C. and about 95° C., for about a half hour while maintaining the mixture at the neutral point or slightly on the alkaline side.

The mixture is then cooled, for example to room temperature to insure relatively complete precipitation of the product, leaving in solution the salts formed in the neutralization step, e. g. alkali metal or ammonium chloride and sulfate.

Separation of the precipitated product may be effected in any desired manner, for example by filtration, centrifugation, etc. and the recovered product may be dried and ground for storage or use. The final product is obtained in hydrate form as a white or light gray solid. The decachlorotetrahydro-4,7-methanoindeneone of our invention may exist in all degrees of hydration from substantially anhydrous to an undried gelatinous solid ostensibly containing 67.3 mols H_2O per mol anhydrous material. A number of examples—chosen at random—are shown in Table I below:

TABLE I

Sample No.	Weight Percent H_2O	Mols H_2O per Mol Anhydrous Compound	Comment
----------	0	0	Theory for anhydrous compound.
----------	3.54	1	Theory for monohydrate.
----------	6.84	2	Theory for dihydrate.
1	4.76	1.36	Dried 18 hours at 90° C.
2	1.70	.47	Dried 72 hours at 90° C.
3	10.90	3.33	Dried 72 hours at 27° C.
4	3.02	.85	----------
5	3.04	.86	----------
6	71.2	67.3	----------
7	3.20	.90	----------
8	.10	.03	----------
9	2.7	.77	----------

In carrying out the process for preparing the decachlorotetrahydromethanoindeneone compound of our invention according to the procedure wherein a halosulfonic acid, such as fluosulfonic acid or chlorosulfonic acid, is used as the condensing agent, hexachlorocyclopentadiene and the halosulfonic acid are mixed in a reaction vessel for a period sufficient to form a solid reaction product of hexachlorocyclopentadiene and halosulfonic acid.

The solid hexachlorocyclopentadiene-halosulfonic acid reaction product thus formed is then hydrolyzed to the decachlorotetrahydromethanoindeneone of our invention in a relatively large volume of an aqueous drowning medium.

The halosulfonic acid used as condensing agent to form the hexachlorocyclopentadiene-halosulfonic acid reaction product may be chlorosulfonic acid or fluosulfonic acid, used either alone,

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or dissolved in a solvent which is inert to the reaction such as tetrachloroethylene.

The temperature of the initial condensation reaction may be somewhat higher than that used when SO_3 is the condensing agent and may be carried as high as 140° C., if desired, and thus effect a reduction in the time necessary for completion of the reaction over that necessary when lower temperatures are employed. Usually temperatures between about 90° C. and about 120° C. are satisfactory to effect completion of the condensation reaction in from about 5 to about 2 hours, respectively.

The molar ratio of halosulfonic acid to hexachlorocyclopentadiene for good yields of reaction product should be at least about 1:2, and we have found that ratios between about 1.25:2 and about 8:2 are satisfactory.

The hydrolysis of the hexachlorocyclopentadienehalosulfonic acid reaction products to decachlorotetrahydromethanoindeneone may be effected by heating the reaction product with an aqueous caustic alkali solution, a water solution alone being ineffective to accomplish complete hydrolysis, probably because of the low solubilities of these reaction products in water. In the case of the chlorosulfonic acid product, hydroysis may be carried out in aqueous lower aliphatic alcohol solution which appears to impart sufficient solubility to the reaction product to promote hydrolysis. However, the fluosulfonic acid reaction product requires at least some alkali in the solution for adequate hydrolysis. Temperatures at which the hydrolyses are carried out are not particularly critical, but preferably should be at least about 30° C., and temperatures between about 60° C. and about 90° C., that is, temperatures in the vicinity of the boiling points of the lower aliphatic alcohols are satisfactory.

In general, no digestion step is necessary following the hydrolysis of the hexachlorocyclopentadienehalosulfonic acid reaction products as the decachlorotetrahydromethanoindeneone product precipitates readily from the reaction mixture upon acidification with aqueous mineral acids.

The toxicity characteristics of our new decachlorotetrahydro-4,7-methanoindeneone (designated DTMO in the tables) against various pest organisms are indicated by the following tests, results of which are tabulated below.

Comparative tests were run with DDT and the new compound of our invention (DTMO) on laboratory reared DDT-resistant houseflies, with results as shown in Table II. Tests were run by spraying the flies with a solution of toxicant in spray oil ("Ultrasene" brand deodorized kerosene).

TABLE II

Toxicity of DTMO to houseflies vs. DDT

Treatment	Alive		Dead		Percent Dead	
	Males	Females	Males	Females	Males	Females
DTMO at 0.25%	5	89	56	0	91.8	0.0
DTMO at 0.50%	0	19	52	53	100.0	73.6
DDT at 0.25%	27	69	18	3	40.0	4.2
DDT at 0.50%	8	39	53	3	86.9	7.1
Check (no toxicant)	20	29	0	0	0.0	0.0

Our new compound was compared with 2,3,4,5,-6,7,8,8-octachlorotetrahydro-4,7-methanoindane ("Chlordane") against flies in tests in which panels were sprayed with solutions of the toxicants, with results shown in Table III.

TABLE III

Toxicity of DTMO to houseflies vs. "Chlordane"

Treatment	Spraying Time of Panels (sec.)	No. Flies	No. knocked down after—		
			8 hrs.	24 hrs.	32 hrs.
DTMO, 5%	5	33	5	32	33
Isopropanol, 25%	10	32	7	31	32
Ultrasene, 70%	15	31	17	31	......
Chlordane, 5%	5	26	17	22	25
Isopropanol, 25%	10	35	31	33	34
Ultrasene, 70%	15	33	33	......	......
Check (no toxicant)	....	32	0	0	0

The tests recorded in Table IV compare DTMO with DDT (both as 5% dusts) against southern armyworms and Mexican bean beetle larvae.

TABLE IV

Toxicity of DTMO to armyworms and bean beetles vs. DDT

Treatment	Percent Kill Against—	
	Armyworms	B. B. Larvae
DTMO, 3%; Clay, 97%	100	80.0
DDT, 3%; Clay, 97%	100	66.6

Results on American cockroaches are shown in Table V below, using two hundred mg. of dust for each test.

TABLE V

Toxicity of DTMO to cockroaches

Treatment	No. Roaches	No. knocked down after—			
		6 hrs.	20 hrs.	28 hrs.	48 hrs.
DTMO, 5%; Clay, 97%	10	0	0	5	10
Check (no toxicant)	5	0	0	0	0

Results on stored grain insects, i. e. bean weevil and confused flour beetle are shown in Table VI.

TABLE VI

Toxicity of DTMO to grain insects

Treatment	Percent Kill—		
	Hours	Bean Weevil	Confused Flour Beetle
DTMO, 1.25% in flour	24 48	20 80	10 90
DTMO, 2.50% in flour	24 48	100 100	100 100
DTMO, 5.00% in flour	24 48	100 100	100 100
Chlordane, 1.25% in flour	24 48	20 90	0 80
Chlordane, 2.50% in flour	24 48	100 100	100 100
Chlordane, 5.00% in flour	24 48	100 100	100 100
Check (no toxicant)	48	0	0

Carpet beetle larvae are customarily used in evaluating moth-proofing characteristics of wool-

DEFENDANT'S EXHIBIT 5

ens since they are larvae of the same general type as those of the clothes moth and they not only are in general significantly more resistant to toxicants than the clothes moth larvae, but are actually responsible for a large proportion of the damage to woolen materials loosely characterized as moth damage. "Moth-proofing" tests using carpet beetle larvae as test organisms were carried out by soaking woolen swatches in 1% acetone solution of our decachlorotetrahydromethanoindeneone and "DDT" for comparison. The swatches were dried, weighed to determine deposit, dry cleaned as indicated to determine the tenacity of its adherence to cloth, and exposed to larvae before and after dry cleaning, with results shown in Table VII.

TABLE VII

Toxicity of DTMO to carpet beetles vs. DDT

Treatment	Percent Deposit on Swatch	No. Dry Cleanings	Percent Deposit After Dry Cleaning	No. Larvae	Observations After Three Weeks	
					Dead	Feeding
DTMO, 1% in Acetone:						
1	2.8	2	2.1	20	20	Trace.
2	2.5	2	1.8	20	20	Do.
3	2.2	1	2.2	20	20	None.
4	2.4	1	2.3	20	19	Trace.
5	2.8	0	----------	20	20	None.
DDT, 1% in Acetone:						
6	2.7	2	1.7	20	7	Trace.
7	2.5	2	1.9	20	13	Do.
8	2.5	1	1.6	20	5	Do.
9	3.0	1	1.8	20	14	Do.
10	2.3	0	----------	20	20	None.

It is evident from the results shown in Table VII that not only is our compound more destructive to the carpet beetle larvae than is DDT, but that it is more resistant than DDT to dry cleaning, being an effective toxicant even after at least two cleanings.

The prolonged insecticidal effectiveness of our compound is indicated by the results recorded in Table VIII. In the reported tests, bean plants were dusted with 3% dusts of our compound and of "Chlordane" respectively, then infested with 6th instar southern armyworms at the time intervals indicated.

TABLE VIII

Residual insecticidal activity of DTMO vs. Chlordane

	Age of Deposit	Percent Kill
DTMO, 3% Dust	1 hr.	100
	8 days	100
	10 days	100
	13 days	100
"Chlordane," 3% Dust	1 hr.	100
	8 days	50
	10 days	13.3
	13 days	0

Fungicidal properties of our compound against early blight of potatoes (*Alternaria solani*) are shown by the results set forth in Table IX of field tests in which our compound, mixed with a test insecticide, was applied to potatoes in nine spray applications in the standard manner at about two week intervals in comparison with the results obtained with the test insecticide alone.

TABLE IX

Toxicity of DTMO toward fungi

	Percent Early Blight After Treatment	Total Yield, Bu. Per Acre
DTMO [1]+Test Insecticide [2]	16.25	406
Test Insecticide Alone [3]	31.0	358

[1] 25% DTMO+75% clay—5 lb./100 gals. of water.

[2] Test insecticide—(25% dichlorodiphenyltrichloroethane+3% para nitro phenyl diethyl thiono phosphate in a petroleum emulsion) at the rate of 1 lb. of emulsion per 800 gals. of water.

[3] 1 lb./400 gals.

Field tests carried out on a Montana range on which grasshoppers averaged 12–15 per square yard showed the results listed in Table X when the toxicants listed were applied to the range with a power turbine blower at the indicated rate per acre.

TABLE X

Toxicity of DTMO to grasshoppers vs. Chlordane

Insecticide	Pounds Applied Per Acre	Percent Kill	
		24 hrs.	72 hrs.
DTMO	1	60	100
"Chlordane"	1	----------	75

Our compound, mixed with the same test insecticide described in Table IX, was applied to potatoes in New Hampshire fields in comparison with the same test insecticide used alone. The potatoes were given four spray treatments at about two week intervals with each composition. Seven days after the fourth treatment, the insects were dislodged from the plants by the customary "sweep" technique, caught and counted. Table XI shows the relative number of the various insects remaining after the treatments and also shows the improvement in yield of potatoes after treatment with our compound over that when the test insecticide alone was used.

TABLE XI

Toxicity of DTMO to several potato infesting insects

Insecticide	No. of Organisms Caught by Sweeping After Fourth Treatment			Bu. per Acre
	Flea Beetle	Plant Bugs	Leaf Hoppers	
DTMO Comp.[1]+Test Insecticide [2]	5	3	2	304
Test Insecticide [3]	61	15	3	220

[1] 25% DTMO+75% clay—6 lbs./800 gals. of water.

[2] 25% DDT and 3% p-nitrophenyl diethyl thiono phosphate in a petroleum emulsion—1 lb. of emulsion per 800 gals. of water.

[3] As 1 lb./400 gals. of water.

While the above describes the preferred embodiments of our invention, it will be understood that departures may be made therefrom within the scope of the specification and claims.

We claim:

1. A pesticidal composition comprising as its essential active ingredient a decachlorotetrahydro-4,7-methanoindeneone and a pesticidal adjuvant therefor.

2. A pesticidal composition as defined in claim 1 wherein said active ingredient is dissolved in a

DEFENDANT'S EXHIBIT 5

a pesticidal adjuvant comprising a liquid organic solvent for the active ingredient.

3. A pesticidal composition as defined in claim 1 wherein said active ingredient is dissolved in a pesticidal adjuvant comprising a hydrocarbon liquid solvent for the active ingredient.

4. A pesticidal composition comprising as its essential active ingredient a decachlorotetrahydro-4,7-methanoindeneone admixed with a water-insoluble solid diluent as pesticidal adjuvant therefor, both in finely divided form.

5. A pesticidal dusting powder comprising a minor proportion of a finely divided decachlorotetrahydro-4,7-methanoindeneone as its essential active ingredient admixed with a major proportion of finely divided clay as pesticidal adjuvant therefor.

6. A method for combating noxious pest organisms which comprises contacting said organisms with a composition containing as an essential active ingredient a decachlorotetrahydro-4,7-methanoindeneone.

7. A method for combating insects of the hymenoptera species which comprises contacting said insects with a composition containing as an essential active ingredient a decachlorotetrahydro-4,7-methanoindeneone.

8. A method for combating insects of the lepidoptera species which comprises contacting said insects with a composition containing as an essential active ingredient a decachlorotetrahydro-4,7-methanonideneone.

9. A method for combating insects of the coleoptera species which comprises contacting said insects with a composition containing as an essential active ingredient a decachlorotetrahydro-4,7-methanoindeneone.

10. A method for combating insects of the orthoptera species which comprises contacting said insects with a composition containing as an essential active ingredient a decachlorotetrahydro-4,7-methanoindeneone.

11. A method for combating fungi which comprises contacting said fungi with a composition containing as an essential active ingredient a decachlorotetrahydro-4,7-methanoindeneone.

EVERETT E. GILBERT.
SILVIO L. GIOLITO.

REFERENCES CITED

The following references are of record in the file of this patent:

UNITED STATES PATENTS

Number	Name	Date
2,507,207	Hyman	May 9, 1950
2,519,190	Hyman	Aug. 15, 1950

OTHER REFERENCES

Newcomer et al.: J. Am. Chem. Soc., volume 71, pages 946, 951, March 1949).

Certificate of Correction

Patent No. 2,616,825 November 4, 1952

EVERETT E. GILBERT ET AL.

It is hereby certified that error appears in the printed specification of the above numbered patent requiring correction as follows:

Column 1, line 31, for "adayted" read *adapted*; line 51, in the table, third column thereof, for "296" read *496*;

and that the said Letters Patent should be read as corrected above, so that the same may conform to the record of the case in the Patent Office.

Signed and sealed this 7th day of April, A. D. 1953.

[SEAL]

THOMAS F. MURPHY,
Assistant Commissioner of Patents.

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[Filed June 6, 1958]

UNITED STATES DISTRICT COURT
FOR THE DISTRICT OF COLUMBIA

PURDUE RESEARCH FOUNDATION :
Plaintiff :
:
v. : Civil Action 714-56
:
ROBERT C. WATSON :
Commissioner of Patents :
Defendant :

MEMORANDUM

This is an action under 35 U.S.C. §145 to authorize the Commissioner of Patents to issue to the plaintiff letters patent containing Claims 5, 6, 7, 8 and 13 on an application of Earl T. McBee and Jack S. Newcomer, Serial No. 238,186 filed on July 23, 1951. There is no prior art relied on and the decision of the Board of Appeals of the Patent Office refused to allow the claims on the sole ground that they do not adequately define the invention in accordance with 35 U.S.C. §112, which, so far as pertinent here, reads as follows:

> "The specification shall conclude with one or more claims particularly pointing out and distinctly claiming the subject matter which the applicant regards as his invention."

The Court is of the opinion that the "fingerprint characteristics" which define the compound in Claim 6 do not adequately and distinctly define the chemical compound as required by the statute as quoted, and that Claim 13 which also contains a reference to a process by which the compound is preparable similarly fails to meet the requirements of the statute.

It was agreed at trial that disposition of Claims 5, 7 and 8 would be controlled by disposition of Claim 6.

The Court is of the opinion that the decision of the Board of Appeals of the Patent Office has a reasonable basis and is correct.

Accordingly, the complaint filed herein will be dismissed.

Counsel for the defendant will submit proposed findings of fact, conclusions of law and judgment consistent with the foregoing.

/s/ Joseph C. McGarraghy
Judge

June 6, 1958

[Filed June 26, 1958]

UNITED STATES DISTRICT COURT
FOR THE DISTRICT OF COLUMBIA

PURDUE RESEARCH FOUNDATION,
Plaintiff,

v.

ROBERT C. WATSON,
COMMISSIONER OF PATENTS,
Defendant.

CIVIL ACTION
No. 714-56

JUDGMENT

This action came on to be heard at the January term last, and thereupon upon consideration thereof, it is this 26 day of June, 1958

ADJUDGED that the complaint be and it is hereby dismissed, with costs against plaintiff.

/s/ Jos. C. McGarraghy
Judge

APPROVED AS TO FORM:

/s/ Dean Laurence
Attorney for Plaintiff

[Submitted by Arthur H. Behrens,
Attorney for Defendant]

[Filed June 26, 1958]

FINDINGS OF FACT

1. Purdue Research Foundation, a corporation of Indiana, brought this action under 35 U. S. C. 145 to authorize the Commissioner of Patents to issue letters patent to it containing claims 5, 6, 7, 8 and 13, of the application for patent of Earl T. McBee and Jack S. Newcomer entitled "Composition of Matter," Serial No. 238,186, filed July 23, 1951. Purdue Research Foundation assertedly is the assignee of the McBee and Newcomer application.

2. The application claims 5, 6, 7, 8, and 13, in suit here, are directed to a chemical compound. Claim 6 is representative of claims 5, 6, 7, and 8.

3. Claim 6 reads as follows:

> A composition of matter which has the following characteristics: it is a white crystalline solid having a sealed tube corrected melting point of about 355 degrees Centigrade, it behaves as a weak organic acid, is very soluble in polar organic solvents, substantially insoluble in non-polar organic solvents, and has the approximate molecular formula $C_{10}Cl_{10}O$.

4. Claim 13 reads as follows:

> A composition of matter which has the following characteristics: it is a white crystalline solid having a sealed tube corrected melting point of about 355 degrees Centigrade, it behaves as a weak organic acid, is very soluble in polar organic solvents, substantially insoluble in non-polar organic solvents and has the approximate formula $C_{10}Cl_{10}O$ and is preparable from a compound having a melting point of approximately 146-147 degrees Centigrade, formed by the reaction of hexachlorocyclopentadiene with chlorosulfonic acid, by treatment with alcoholic alkali and acidification and dehydration.

5. Counsel for the plaintiff and defendant agreed at trial that claims 5, 7, and 8 should stand or fall with claim 6. Counsel for plaintiff and defendant agreed also that if the Court should allow either claim 6 or claim 13 the other claim, claim 13 or claim 6, should not be allowed.

6. Claims 5, 6, 7, 8, and 13 were rejected on the ground that these claims do not adequately define the invention in accordance with 35 U. S. C. 112, second paragraph, because they do not particularly point out and distinctly claim the compound which the applicant regards as his invention.

7. The McBee and Newcomer application is directed to a chemical compound which is said to be useful in its anhydrous form as a dessicant because of its hygroscopic nature, and as an ingredient in insectidical compositions. The application as filed stated that this compound was "believed to have the approximate molecular formula $C_{10}HCl_{10}O_2$" and that "the exact elemental composition and structure of the compound are not precisely known at the present time." Over a year after the filing of the application it was amended to change the "approximate molecular formula" of the compound from "$C_{10}HCl_{10}O_2$" to "$C_{10}Cl_{10}O$," which change involved the elimination of one hydrogen atom and one oxygen atom from the formula, and to modify the specification to state that the "molecular structure" (instead of the "elemental composition and structure") is "not precisely known." Subsequent to the final rejection of the application McBee and Newcomer filed a supplementary oath which was allegedly applicable to the subject matter of their claims which was disclosed but not claimed in their application as originally filed in the Patent Office.

8. The specification of the McBee and Newcomer patent application states that the composition of the present invention exists in the anhydrous form as well as in the form of various hydrates and that the anhydrous and hydrate forms all give the same melting point in a sealed tube of approximately 355 degrees Centigrade, corrected. A tabulation, in the specification, of melting point data for "$C_{10}HCl_{10}O_2$" (later amended to read $C_{10}Cl_{10}O$) shows a variation of melting points (degrees Centigrade corrected) from 331 to 350 at "1st Soft;" from 351 to 355 at "1st Meniscus;" and from 351 to 361 at "1st Crystal."

9. Plaintiff's expert witness, James S. Sconce, testified that an analytic chemist accepts as a melting point range, as a rule, the zone between the first crystal point and the last crystal point, and that in his laboratory melting points were determined, for substances melting in the neighborhood of 20 to 50 degrees Centigrade, accurately to a hundredth of one degree. He testified that for substances melting as high as 355 degrees Centigrade a much wider figure, two degrees, three degrees, or four degrees, was "significant."

10. Textbooks show, for pure substances the change from the solid state to the liquid state is quite sharp (within 0.5°C.), hence this temperature, the melting point, is valuable for purposes of identification. It is in many cases sufficient to determine the melting point with an accuracy of 0.5 to 1°C. Reported discrepancies in melting point determinations amounting to more than 2°C. with different types of apparatus and as much as 1°C. with different observers using the same apparatus represent the maximum amount of divergence. The melting point of the claimed compound is in a range where the melting points can not be determined with the indicated degree of accuracy. The reported melting point of plaintiff's claimed compound, which is subject to decomposition at temperatures near its melting point, was not determined with a degree of accuracy to make it, by the standards of the organic chemistry art, an identifying characteristic.

11. As the Board's decision states, a molecular formula represents a stated number of atoms of each of the kinds (elements) included in the formula so that a limiting term like "approximate" as applied to the molecular formula of claim 6 (representative of claims 5, 6, 7, and 8) could only have significance as indicating that the formula may be varied by a whole number of any one or more of the three kinds of atoms indicated. For this reason the molecular formula as qualified by "approximate" is meaningless as an item of identification of the claimed chemical compound.

12. The molecular formula specified in the claims of the McBee and Newcomer application is not derivable from the McBee and Newcomer application as originally filed so it cannot reasonably be asserted that the claimed formula constitutes essential identifying information.

13. The general statement of solubility in claim 6, that the compound is "very soluble in polar organic solvents" and "substantially insoluble in non-polar organic solvents," and the assertion in claim 6 that the compound "behaves as a weak organic acid" are so broad as to obviously apply to many compounds of entirely different characteristics.

14. The monohydrate of the organic compound of the claims in suit is described in the McBee and Newcomer specification as behaving as does a weak organic acid in that it gives an indefinite end-point with phenolphthalein. Weak organic acids, as plaintiff's expert witness testified on cross-examination, have many characteristics in common but differ among themselves in various significant characteristics. The dissociation constant, a measure of the hydrogen ion liberation in a compound in water solution, has a larger value for strong acids than for weak acids but the claims in suit do not specify that the dissociation constant, or any specific item of behavior, shall be the criterion of behavior. The organic compound of McBee and Newcomer is not a weak organic acid. It is now fairly well accepted that this compound is a ketone but the McBee and Newcomer application as filed did not so identify it.

15. Although claim 13 describes the plaintiff's compound by reference to the process by which it is "preparable" in addition to a recitation of its "characteristics," it is not restricted to read on compounds produced by the process referred to in the claims or to compounds produced by processes disclosed in the McBee and Newcomer specification.

16. The "characteristics" of the claimed compound as set forth in claim 6 (representative of claims 5, 6, 7, and 8), which are referred to by plaintiff as the compound's "finger prints," do not, when taken individually or in combination, adequately and distinctly describe the claimed compound. Claim 13 is similarly deficient.

CONCLUSIONS OF LAW

1. Under 35 U. S. C. 112 the claim of an application for patent must particularly point out and distinctly claim the subject matter which the applicant regards as his invention.

2. As provided in Rule 118 of the Rules of Practice Of The United States Patent Office In Patent Cases,

In original applications, all amendments of the drawings or specification, and all additions thereto, must conform to at least one of them as it was at the time of the filing of the application. Matter not found in either, involving a departure from or an addition to the original disclosure, cannot be added to the application even though supported by a supplemental oath, and can be shown or claimed only in a separate application.

3. Where one has produced a composition of matter in which invention rests over prior art compositions, and where it is not possible to define the characteristics which make it inventive except by referring to the process by which the composition is produced, one is permitted to so claim his composition, but his protection is limited to compositions produced by his process referred to in the claims. When the composition is thus claimed in terms of the process of preparation, the product cannot be defined in such a manner as to assert a monopoly on the product by whatever means produced.

4. Claims 5, 6, 7, 8, and 13 do not meet the requirements of 35 U. S. C. 112.

5. Plaintiff is not entitled to a patent containing any of the claims 5, 6, 7, 8, and 13 of the McBee and Newcomer application Serial No. 238,186.

The complaint should be dismissed as to all the claims in suit.

/s/ Joseph C. McGarraghy
Judge

June 26, 1958

[Filed August 22, 1958]

NOTICE OF APPEAL TO COURT OF APPEALS UNDER RULE 73(b)

Notice is hereby given that PURDUE RESEARCH FOUNDATION, Plaintiff above named, hereby appeals to the United States Court of Appeals for the District of Columbia Circuit from the final Judgment dismissing the complaint entered in this action on 1958 June 26, and from the whole thereof.

Dated this 22 day of August 1958.

Respectfully,

/s/ Herbert I. Sherman
Herbert I. Sherman
Attorney for Plaintiff
753 Warner Building
Washington 4, DC

Of Counsel:

LAURENCE AND LAURENCE

/s/ Dean Laurence

Copy to be served by mail on Clarence W. Moore, Solicitor, U. S. Patent Office and Counsel for Defendant, Washington 25, DC.

Feb. 25, 1958 E. E. GILBERT ET AL Re. 24,435

PROCESS FOR PREPARING [DECACHLOROTETRAHYDRO-4,7-METHANOINDENEONE] A HYDROLYZED REACTION PRODUCT OF HEXACHLOROCYCLOPENTADIENE AND SULFUR TRIOXIDE

Original Filed Nov. 17, 1950 2 Sheets-Sheet 1

[DECACHLOROTETRAHYDRO-4,7-METHANOINDENEONE]

KETONIC $C_{10}Cl_{10}O$ COMPOUND

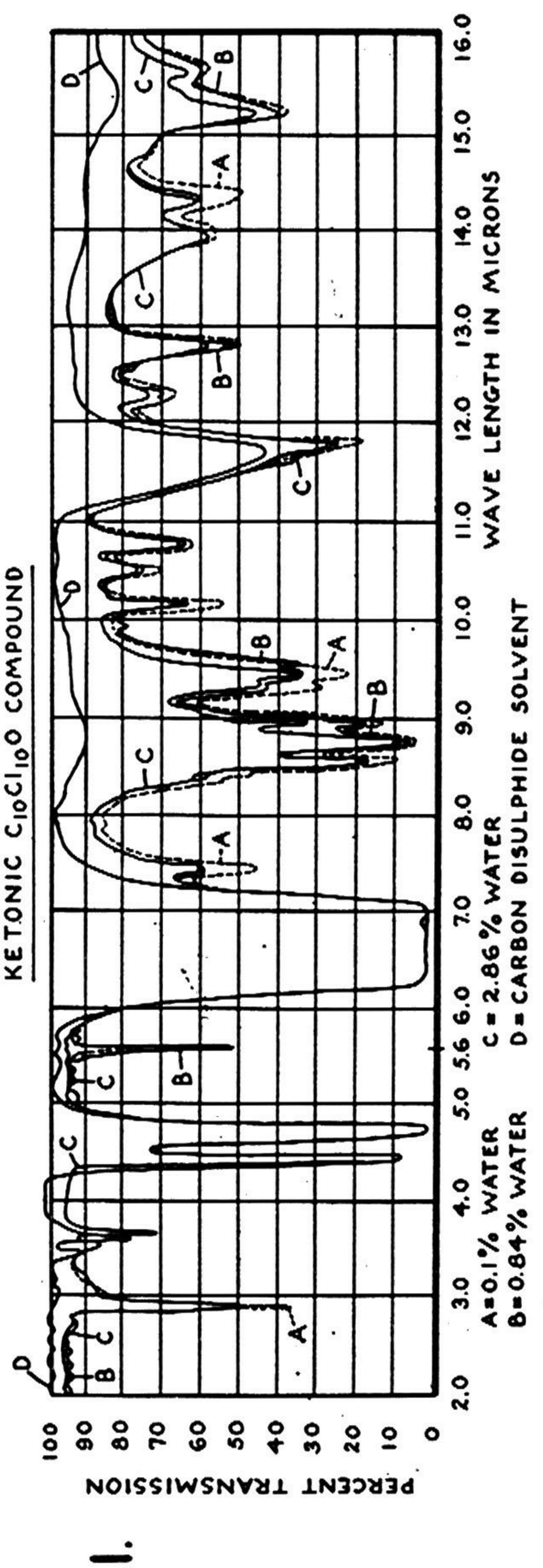


FIG.1.

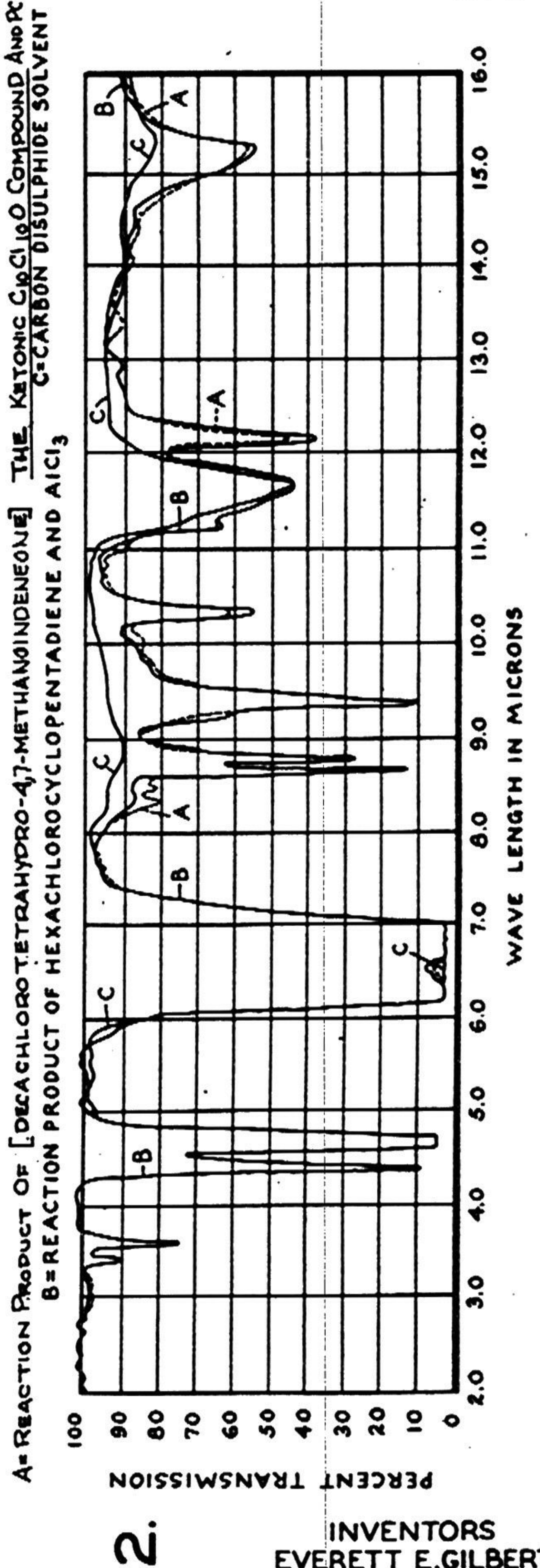


FIG.2.

INVENTORS
EVERETT E. GILBERT
SILVIO L. GIOLITO
BY
Elizabeth Hunter
ATTORNEY

Feb. 25, 1958 E. E. GILBERT ET AL Re. 24,435

PROCESS FOR PREPARING [DECACHLOROTETRAHYDRO-4,7-METHANOINDENEONE]
A HYDROLYZED REACTION PRODUCT OF HEXACHLOROCYCLOPENTADIENE
AND SULFUR TRIOXIDE

Original Filed Nov. 17, 1950 2 Sheets-Sheet 2

A=Reaction Product of [Decachlorotetrahydro-4,7-methanoindeneone] The Ketonic $C_{10}Cl_{10}O$ Compound and Acetic Anhydride

B=Carbon Disulphide Solvent

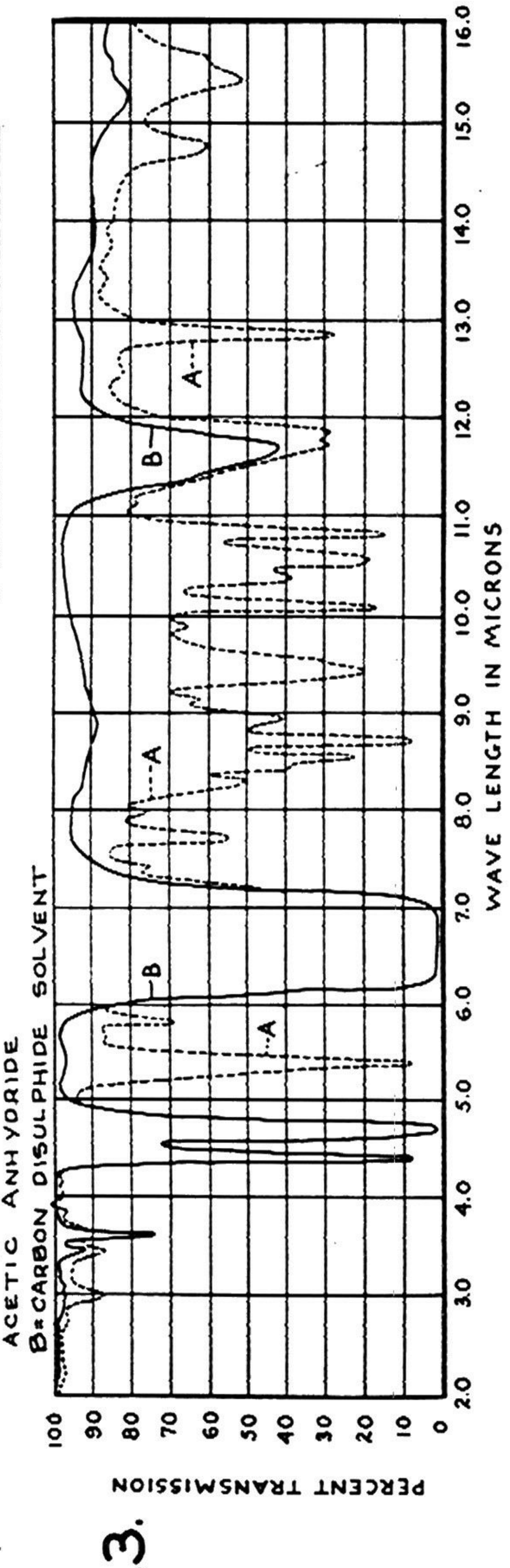


FIG. 3.

A=Reaction Product of [Decachlorotetrahydro-4,7-methanoindeneone] The Ketonic $C_{10}Cl_{10}O$ Compound and Acetic Anhydride with Ethyl Alcohol

B=Carbon Disulphide Solvent

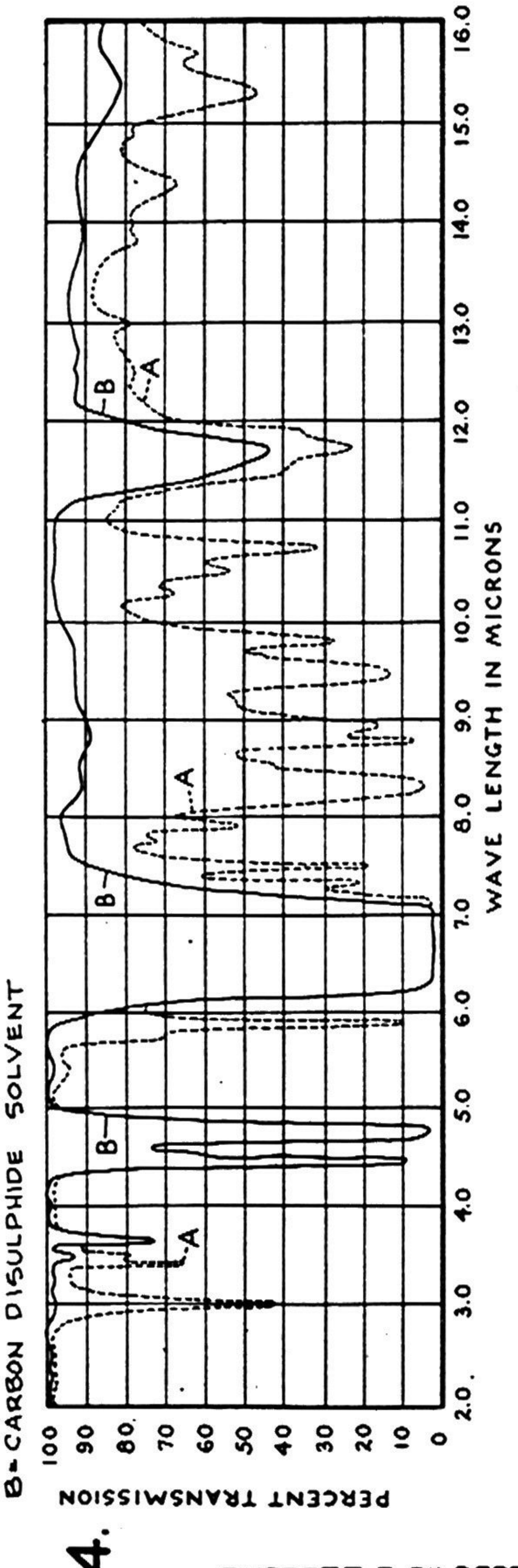


FIG. 4.

EVERETT E. GILBERT
SILVIO L. GIOLITO
BY
Elizabeth Hunter
ATTORNEY

United States Patent Office

Re. 24,435
Reissued Feb. 25, 1958

24,435

PROCESS FOR PREPARING [DECACHLORO-TETRAHYDRO-4,7-METHANOINDENEONE] A HYDROLYZED REACTION PRODUCT OF HEXACHLOROCYCLOPENTADIENE AND SULFUR TRIOXIDE

Everett E. Gilbert, Morris Township, Morris County, N. J., and Silvio L. Giolito, Whitestone, N. Y., assignors to Allied Chemical & Dye Corporation, New York, N. Y., a corporation of New York

Original No. 2,616,928, dated November 4, 1952, Serial No. 196,123, November 17, 1950. Application for reissue August 19, 1957, Serial No. 679,114

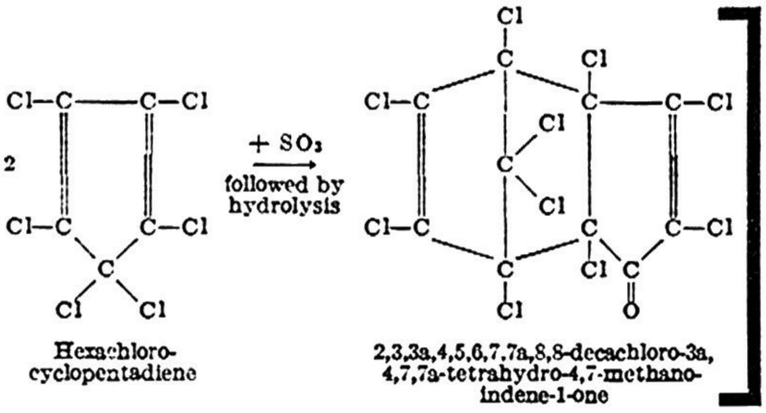
5 Claims. (Cl. 260—586)

Matter enclosed in heavy brackets [ ] appears in the original patent but forms no part of this reissue specification; matter printed in italics indicates the additions made by reissue.

This invention relates to a method for preparing a [decachloro-tetrahydro-4,7-methanoindeneone] *new ketonic compound* by condensing two molecules of hexachlorocyclopentadiene with the aid of sulfur trioxide to form a hexachlorocyclopentadiene-SO_3 reaction product and hydrolyzing the reaction product to the ketone.

The resulting [decachloro - tetrahydro - 4,7 - methanoindeneone] *ketonic compound* is useful as an insecticide, as a fungicide, and as a moth-proofing agent, as disclosed and claimed in co-pending application Serial No. 196,121, filed November 17, 1950, *now U. S. Patent 2,616,825.*

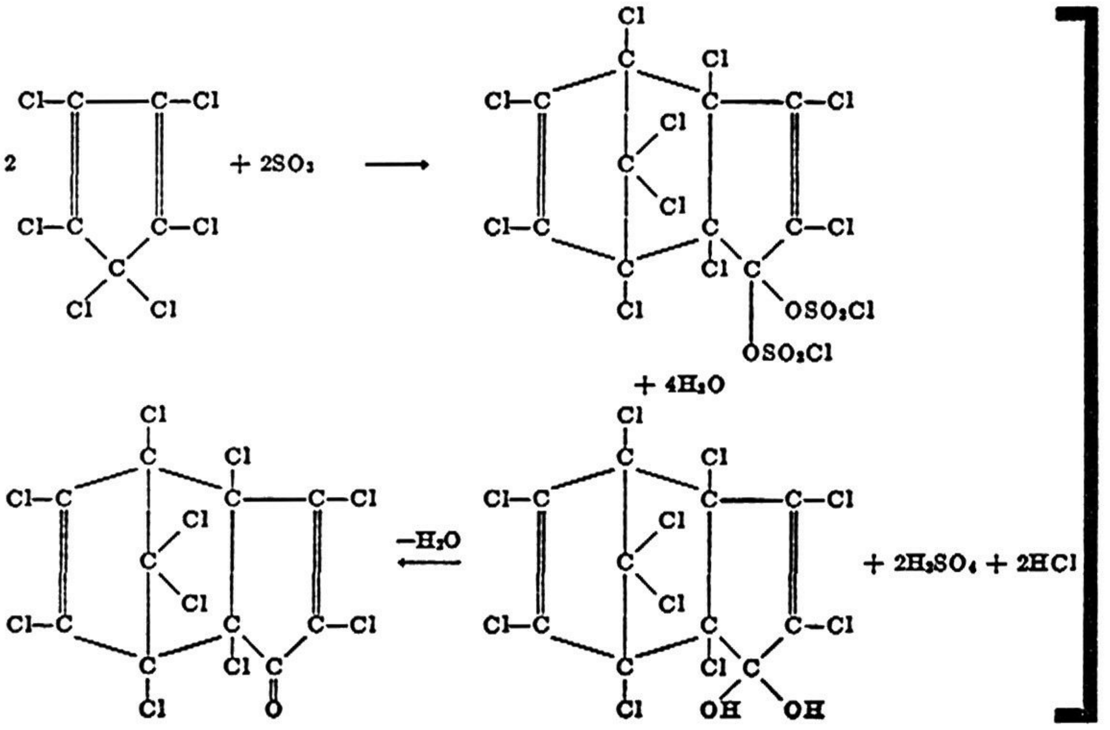
The exact mechanism of the new reaction, particularly in its intermediate stages, is not clearly understood, but the overall reaction *results in a ketonic compound having the empirical formula $C_{10}Cl_{10}O$* [is indicated in the equation set forth below:



Hexachloro-cyclopentadiene

2,3,3a,4,5,6,7,7a,8,8-decachloro-3a,4,7,7a-tetrahydro-4,7-methano-indene-1-one]

[The intermediate stages of the reaction are believed to proceed somewhat as illustrated below:



]

The drawings, Figs. 1–4 inclusive, represent infrared spectrograms of the [decachlorotetrahydro-4,7-methanoindeneone] *ketonic compound* prepared according to our invention, and of certain of its reaction products with other organic compounds.

In the drawings, Fig. 1 represents the infrared spectrogram of the [decachlorotetrahydro-4,7-methanoindeneone,] *ketonic compound,* as obtained in carbon disulfide solution, the several lines showing the spectrograms recorded of samples of different degrees of hydration; dotted line A being the record of substantially anhydrous material (0.10%, 0.03 mol, H_2O); solid line B being the record of a slightly hydrated sample (0.84%, 0.24 mol, H_2O); while solid line C is the record of an essentially monohydrated sample (2.86%, 0.8 mol, H_2O). The three spectrograms are considered substantially identical. Solid line D is the spectrogram of the carbon disulfide solvent. Fig. 2 represents the infrared spectrogram of the reaction product of the [decachlorotetrahydro-4,7-methanoindeneone] *$C_{10}Cl_{10}O$ compound* with PCl_5, shown as broken line A, and of the compound obtained by the reaction of hexachlorocyclopentadiene with $AlCl_3$ by known methods, shown as solid line B, both as obtained in carbon disulfide solution. The two spectrograms are substantially identical (except for minor variations due to small quantities of impurities), hence the products prepared by the two processes are identical. Fig. 3 represents the infrared spectrogram, shown as broken line A, of the reaction product of our new *ketonic* compound with acetic anhydride as described hereinafter. This spectrogram is similarly superimposed on the spectrogram, shown as solid line B, of the carbon disulfide solvent. Fig. 4 represents the infrared spectrogram, shown as broken line A, of the product obtained by reacting our new compound successively with acetic anhydride and then with ethyl alcohol as described hereinafter. This spectrogram is similarly superimposed on the spectrogram of carbon disulfide, used as solvent, and shown as solid line B.

In carrying out our process for preparing the [decachlorotetrahydro - 4,7 - methanoindeneone] *new ketonic* compound above described, hexachlorocyclopentadiene and sulfur trioxide are mixed by charging them, either simultaneously or separately, to a reaction vessel. After mixing the reactants, the charge may be digested for a short period, sufficient to condense two molecules of hexachlorocyclopentadiene and to form a liquid reaction product of hexachlorocyclopentadiene and sulfur trioxide.

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The liquid SO_3-hexachlorocyclopentadiene reaction product thus formed is then hydrolyzed to the [decachloromethanoindeneone] *ketone* by drowning the reaction product in a relatively large volume of an aqueous drowning medium, preferably an alkaline aqueous medium, for example, in at least about 5 volumes of drowning medium per volume of reaction product.

After addition of the reaction product to the drowning medium is complete, the mixture may be digested for a short additional period, for example a half hour, to insure completion of the hydrolysis.

The product may be filtered at this stage, if desired, but to improve coagulation and ease of filtration, we prefer to agitate the batch for a short period at a temperature somewhat higher than the drowning temperature, for example at about 90°–95° C. for about a half hour, during which the crystalline product may partially or completely redissolve.

When the drowning has been effected in an alkaline solution, the charge is then neutralized, for example with sulfuric acid, whereupon the crystalline product reprecipitates. The amount of acid added for neutralization should be sufficient to bring the mixture preferably to neutrality, or to a very slight alkalinity not exceeding about 0.05% NaOH, as indicated by a pH of 7–8.

After neutralization, the charge is agitated, preferably at a temperature between about 90° C. and about 95° C., for about a half hour while maintaining the mixture at the neutral point or slightly on the alkaline side.

The mixture is then cooled, for example to room temperature to insure relatively complete precipitation of the product, leaving in solution the salts formed in the neutralization step, e. g. alkali metal or ammonium chloride and sulfate.

Separation of the precipitated product may be effected in any desired manner, for example for filtration, centrifugation, etc., and the recovered product may be dried and ground for storage or use. The final product is obtained in hydrate form as a white or light gray solid. The [decachlorotetrahydro-4,7-methanoindeneone] *ketone* of our invention may exist in all degrees of hydration from substantially anhydrous to an undried gelatinous solid ostensibly containing 67.3 mols H_2O per mol anhydrous material. A number of examples—chosen at random—are shown in Table I below:

TABLE I

Sample No.	Weight Percent H_2O	Mols H_2O per Mol Anhydrous Compound	Comment
—	0	0	Theory for anhydrous compound.
—	3.54	1	Theory for monohydrate.
—	6.84	2	Theory for dihydrate.
1	4.76	1.36	Dried 18 hours at 90° C.
2	1.70	0.47	Dried 72 hours at 90° C.
3	10.90	3.33	Dried 72 hours at 27° C.
4	3.02	0.85	
5	3.04	0.86	
6	71.2	67.3	
7	3.20	0.90	
8	0.10	0.03	
9	2.7	0.77	

The sulfur trioxide used as the condensing agent may be added in any suitable form, for example as liquid or gaseous SO_3; or dissolved in an inert solvent such as sulfuryl chloride (SO_2Cl_2), or in the form of oleum, preferably a high strength oleum, for example 60% oleum. Mixing of SO_3 and hexachlorocyclopentadiene may be initiated at any convenient temperature, for example room temperature (25° C.) or below. The negative heat of solution of SO_3 in the hexachlorocyclopentadiene may cause momentary temperature drop of a few degrees centigrade, but since the reaction is exothermic, the temperature will usually soon rise spontaneously. If not, the mixture may be heated gently to initiate the reaction. However, during the mixing of SO_3 with the hexachlorocyclopentadiene, the temperature of the charge is preferably maintained at about 35° C. or above to avoid momentarily delaying the sulfonation reaction with the result that the cumulative reaction may take place with violence at a later stage. Furthermore, when using liquid SO_3 as a reactant, it is undesirable to exceed about 45° C. during the course of the addition since liquid SO_3 volatilizes slightly below this temperature. In any event, we prefer to carry out the addition of the reactants at temperatures between about 35° C. and about 70° C. Temperatures above about 70° C. during SO_3 addition appear to cause an increase in the viscosity of the mixture and to promote undesired side reactions which decrease the yield of the desired product. The reactants may be mixed in any desired manner and in any order. However, when operating on a large scale, it is sometimes desirable to mix the reactants step-wise to aid in controlling the temperature of the exothermic reaction, especially when using liquid SO_3. In such cases it is convenient to add the more volatile SO_3 (B. P. 43° C.) to the liquid hexachlorocyclopentadiene gradually or portionwise. A digestion step at a temperature above the reaction temperature, for example about 80° C., may be carried out, if desired, following completion of mixing of the reactants.

The ratio of SO_3 to hexachlorocyclopentadiene for good yields of reaction product should be at least 1:1, and we have found that a slight molar excess of SO_3 over hexachlorocyclopentadiene, for example 1.5:1, improves the yield, and eliminates unreacted hexachlorocyclopentadiene residue in the charge which otherwise would have to be removed from the charge by a separate operating step, such as, for example, steam distillation.

The hydrolysis of the hexachlorocyclopentadiene-SO_3 reaction product may be effected, if desired, by simply drowning the mass in water. However, the extremely corrosive nature of the spent acid thus obtained, containing both sulfuric and hydrochloric acids, makes such solutions extremely difficult to handle in metallic equipment. Accordingly, we prefer to carry out the drowning step in a weak alkali solution, for example a weak alkali metal hydroxide solution. Ammonia or the alkali metal carbonates may be used if desired, but are not as satisfactory as the alkali metal hydroxides as they tend to cause foaming of the charge and sometimes gelling of the precipitated product, rendering it difficult to separate or filter. The use of alkali metal hydroxide concentrations above about 10%, however, appears to cause considerable thickening upon neutralization. Accordingly, we prefer to employ alkali metal hydroxide solutions of not more than about 10% concentration by weight, for example concentrations of between about 6% and about 8% being satisfactory. The aqueous drowning solution should preferably be warm, for example at least about 40° C. at the start of the drowning operation. The charge of reaction product is added rather slowly to the drowning medium, and the temperature rises somewhat due to the exothermic nature of the reaction. When the temperature has risen to about 60°–70° C., external cooling is provided and the remainder of the charge added, preferably while maintaining the temperature between about 60° C. and about 70° C. Upon addition of the reaction product to the drowning medium, droplets usually form which gradually whiten in color due to the formation of crystalline [decachloromethanoindeneone] product; and these droplets gradually disperse to liberate a fine white crystalline precipitate.

As pointed out above, it is desirable to follow the drowing step with a digestion step of short duration, for example between about 30 and about 60 minutes, at an elevated temperature, for example 90°–95° C., to insure complete hydrolysis.

In general, the temperature during the drowning operation appears to determine to a considerable extent the physical character of the resulting precipitated product, a digestion period at an elevated temperature, e. g. ca. 90°–95° C., both before and after neutralization aiding in the formation of a crystalline product which is readily separated from the solution, whereas drowning at lower temperatures, for example 50°–60° C. without a digestion step at the higher temperature indicated above, results in a gelatinous precipitate which is difficult to separate from the reaction mixture. Accordingly, we prefer to carry out the drowning and neutralizing operation under approximately the temperature-time conditions illustrated in the schedule set out in Table II below:

TABLE II

Drowning conditions (6% NaOH soln.)

Step	Time (min.)	Temp. ° C.	Control by—
Addition of Adduct	30–60	50–70	Cooling.
Cooking	30	60–70	Cooling and/or heating.
Heating to 95° C	30	70–95	Heating.
Cooking	30	90–95	Do.
Neutralization	30	90–95	Cooling and/or heating.
Cooking	30	90–95	Heating.
Total Time hours	3–3½	---------	
Cooling	---------	90–30	Cooling.

In order to identify the compound prepared according to our invention and others described herein, their infrared spectrograms were measured and recorded.

The infrared spectrograms shown in the figures were prepared on a standard infrared recording spectrophotometer designed for measuring and recording the infrared transmission of solids, liquids and gases, comprising a double infrared beam which scans the spectrum through the wave length range 2.0 to 16 microns, one part of the beam passing through the sample under study, the other passing through a compensating cell. If the sample under study absorbs radiation, the two beams become unequal. The magnitude of this inequality is a measure of the transmission by the sample of the particular wave length, and the record of these differences within the range of wave lengths scanned is the infrared spectrogram, recorded as an ink drawn line on a chart graduated in percent transmission as ordinates and in wave length as abscissae.

Solid samples, such as the compound of our invention and the related compounds described, are conveniently measured in solution. The spectrograms shown in the figures were all measured by dissolving 0.5 gram of the solid in carbon disulfide and diluting to 10 ml. with the solvent. A small amount of the solution was then introduced into a liquid cell with sodium chloride windows and sealed. The cell was placed in the spectrophotometer in the path of one of the beams as described above.

The infrared spectrogram of any chemical compound serves as an accurate means for identifying the compound. It has been compared with a human fingerprint in its ability to identify a compound with certainty. The characteristic reproducibility of the infrared spectrogram of a given compound is due to the facts that when a molecule is excited by infrared radiation it absorbs energy to a greater degree at some wave lengths than at others, and that the amount of absorption depends on the configuration and upon the linkages of the atoms composing the molecule. Accordingly, the compound prepared according to our invention, and also the other compounds shown in the several figures, are identified and characterized with certainly by their individual spectrograms. The spectrogram of the [decachlorotetrahydro-4,7-methanoindeneone] *ketonic $C_{10}Cl_{10}O$ compound* prepared according to our invention, which, as a solid, exists in various degrees of hydration as pointed out above, is the same regardless of the degree of hydration. This sameness may be explained by the assumption that when the sample is dissolved in the hydrophobic carbon disulfide solvent, most of the water of hydration is disassociated from the compound so that what is measured is the spectrogram of the product itself or of a hydrate containing only a small amount of water. Regardless of any explanation, the characteristic spectrogram persists and is independent of the degree of hydration (while the three spectrograms of the [decachlorotetrahydro - 4,7 - methanoindeneone] *ketonic $C_{10}Cl_{10}O$* compound shown in Fig. 1 exhibit slight variations, these variations are not considered significant).

The following specific examples further illustrate the invention. Parts are by weight except as otherwise indicated.

EXAMPLE 1

A charge of 188 parts (.69 mol) of hexachlorocyclopentadiene was cooled to 5°–10° C., and to the agitated charge was added gradually 940 parts of 60% oleum (containing 565 parts (7.1 mols) of free SO_3). After addition of all the oleum, which required about one hour, the mixture, whose temperature had risen progressively to about 70° C., was added slowly to a large volume (5000 parts) of water to dilute the acid. The crude [decachloromethanoindeneone] *ketonic product* precipitated immediately, upon contact of the charge with the water, as a white solid. The product was filtered from the spent acid, stirred three times with fresh water, and filtered after each water wash, to remove most of the sulfuric acid. The product was further purified by dissolving it in 500 parts 95% ethanol, reprecipitating by the addition of 500 parts water, filtering and drying. 126 parts of purified [decachlorotetrahydro-4,7-methanoindeneone] *$C_{10}Cl_{10}O$ ketone* hydrate were obtained representing a yield of 72% of theoretical.

EXAMPLE 2

240 parts (3 mols) of liquid SO_3 were added rapidly to a stirred solution of 818.1 parts (3 mols) of hexachlorocyclopentadiene and 563 parts (4.17 mols) of sulfuryl chloride (SO_2Cl_2) which was at room temperature. With the addition of the SO_3, the charge temperature rose from 25° C. to 30° C. The charge was then heated gently for four hours at 80° C., then cooled and added with stirring to 5000 parts of water, upon which a white solid precipitated. The mixture was stirred for a half hour, filtered, and twice again mixed with water, stirred and filtered. The solid precipitated product was dissolved in about 2000 parts of methyl alcohol, decolorized with active carbon, the clarified solution drowned in water to precipitate the product, which, upon filtration and drying, yielded 532 parts of [decachlorotetrahydro-4,7-methanoindeneone] *$C_{10}Cl_{10}O$ ketone* hydrate, corresponding to a yield of 70% of theoretical.

EXAMPLE 3

To 50 parts (0.18 mol) of hexachlorocyclopentadiene at room temperature (25° C.) was added dropwise with stirring 14.7 parts (0.18 mol) of liquid SO_3 over a period of about 15 minutes. After the SO_3 had been added, the charge was heated to 80° C. for four hours on a water bath. Then the reaction mixture, a dark red viscous liquid, was poured slowly with stirring into 500 parts of cold water. A white flocculent precipitate formed which was filtered and washed several times with water. This solid reaction product was worked up as described in previous examples to give a final yield of 24.1 parts of [decachlorotetrahydro-4,7-methanoindeneone] *$C_{10}Cl_{10}O$ ketone* hydrate, corresponding to 53% of theoretical.

EXAMPLE 4

273 parts (1.0 mol) of hexachlorocyclopentadiene and 120 parts (1.5 mols) of liquid sulfur trioxide were charged simultaneously to a reaction vessel at room temperature (about 25°) while agitating. Upon mixing, the temperature dropped momentarily an increment of about

5° C. due to negative heat of solution. The temperature then rose spontaneously to 40° C. due to heat of reaction. The reaction temperature was maintained between about 40° C. and about 45° C. for a two hour period while continuing the agitation and while cooling the charge with cooling water circulated through an external water jacket. Following the reaction period, the temperature was gradually raised over a three hour period from 45° C. to 65° C. by external heating. The mixture remained an oily mass throughout the reaction period, but became more viscous as the reaction proceeded, the color changing to a dark red. The mixture was then cooled to room temperature and yielded 390 parts of hexachlorocyclopentadiene-SO_3 reaction product, which was dark red in color and had the consistency of molasses.

The hexachlorocyclopentadiene-SO_3 reaction product was hydrolyzed by slowly pouring the 390 parts of reaction product, obtained as described in the previous paragraph, into an aqueous alkaline solution containing 3100 parts of water and 200 parts of sodium hydroxide (a 6% solution) at a temperature of 40° C. to 50° C. The addition of reaction product consumed about half an hour. A mildly exothermic reaction ensued, the heat of which was soon dissipated by the large volume of the alkaline solution. When about half of the reaction product had been added, the temperature had risen to 50° C. to 60° C. and thereafter cooling was applied during the remainder of the addition to maintain the temperature between 60° C. and 70° C. The thick droplets which formed upon drowning the reaction product, slowly whitened in color and gradually dispersed to fine white particles. After addition of the reaction product was complete, the slurry mixture was digested for about one-half hour while continuing the agitation and while maintaining the temperature between 60° and 70° C. The temperature of the mixture was then raised and maintained between 90° C. and 95° C. for an additional half hour in order to insure complete hydrolysis, whereupon considerable of the crystalline product redissolved in the alkaline solution. Then the mass was neutralized with sulfuric acid by slowly adding over a one hour period, 20 parts of H_2SO_4 as 100% to a very slight alkalinity not exceeding about 0.05% NaOH, as indicated by a pH of 7–8. During the course of the neutralization, some of the [decachlorotetrahydromethanoindeneone product] $C_{10}Cl_{10}O$ *ketone* separated in gelatinous form making continued agitation difficult. However, upon continuing the agitation, the neutral mixture began to coagulate and soon the mixture again became white in color due to separation of the solid product. Agitation of the mixture was continued at 90° C. for an additional half hour while maintaining the pH value at the above figure. The mixture was then cooled to room temperature (about 25° C.) to complete crystallization of the [decachlorotetrahydromethanoindeneone] $C_{10}Cl_{10}O$ *ketone* hydrate. The crystalline product was separated by filtration which was completed in about 10 minutes. The filter cake was washed with water until the filtrate was clear and colorless. The wet filter cake, amounting to 800 parts, was dried in an oven at 110°–115° C. requiring about 36 hours to drive off the water and to bring the cake to constant weight. The dry filter cake amounted to 218 parts corresponding to a yield of [decachlorotetrahydromethanoindeneone] $C_{10}Cl_{10}O$ *ketone* hydrate of 86% of theoretical. The dry product was cooled to 50° C. and pulverized.

The products obtained as described in each of the above examples, had infrared spectrograms substantially identical with those illustrated in Fig. 1 of the drawings.

A portion of [decachlorotetrahydro-4,7-methanoindeneone] $C_{10}Cl_{10}O$ *ketone* hydrate obtained as described in the foregoing examples was purified and dehydrated by repeated vacuum sublimation and analyzed. It had

the following analysis as compared to theoretical for $C_{10}Cl_{10}O$.

	Percent Theory ($C_{10}Cl_{10}O$)	Percent Found
Cl	72.3	72.7
C	24.4	23.8
H	0	0.6
H_2O	0	0.1
Mol Wt	491	496

The [decachlorotetrahydro-4,7-methanoindeneone] $C_{10}Cl_{10}O$ *ketone* hydrate prepared according to our invention is a white crystalline solid with no appreciable odor. Upon heating in a glass melting point tube by conventional procedures up to 300° C., no tendency to melt is noted. It sublimes when heated in the atmosphere; for example, slight sublimation occurs upon oven drying at 110°–115° C. while upon heating at 140° C. at 1–1.5 mm. of Hg pressure, 10–15% of its weight sublimes in three hours. It sublimes with some decomposition when heated in the open atmosphere to 300° C. It is readily soluble in actone, lower aliphatic alcohols, ethers and the like, and also in nitrobenzene and sulfuryl chloride. It is somewhat less soluble in benzene, toluene, hexane and petroleum ether, but is sufficiently soluble in warm hexane to allow the use of this material as a recrystallizing solvent if desired. It is virtually insoluble in cold water and only slightly soluble (less than 0.4%) in boiling water. It tends to gel upon separation by cooling, from hot solutions in hydrocarbon solvents. A satisfactory recrystallizing solvent is 85%–90% aqueous ethanol from which gelation does not occur. It usually exists as a crystalline hydrate when exposed to atmospheric conditions and is useful for insecticidal, etc., purposes in hydrate form. The compound prepared according to our invention is soluble in, and relatively stable toward, strong caustic solutions such as sodium, potassium, and calcium hydroxides. It is identified with certainty by the infrared spectrogram shown in Fig. 1.

The stability towards caustic materials of the compound prepared according to our invention is of advantage in the use of the compound as a pesticide in combination with lime and other alkaline agricultural chemicals, and in this respect it is superior to benzene hexachloride (BHC) and dichlorodiphenyltrichloroethane (DDT) which decompose readily in contact with alkalis. Furthermore, the compound prepared according to our invention, being volatile under normal atmospheric conditions only at temperatures considerably above those usually encountered in use, has a high residual insecticidal activity, and because of its limited solubility in hydrocarbon solvents is considerably more resistant to dry cleaning than DDT, which is an advantage in the use of the compound as a moth-proofing agent.

[The compound prepared according to our invention is believed to be the 2,3,3a,4,5,6,7,7a,8,8-decachloro-3a,4,7,7a-tetrahydro-4,7-methanoindene-1-one, illustrated in the equations set forth above.]

The method of preparation and the reactions of the compound are consistent with the ketonic structure—namely, reaction with phosphorus pentachloride, with butyric anhydride, with acetic anhydride and the further reaction of the acetic anhydride reaction product with ethyl alcohol, and formation of a phenylhydrazone. [Strong indications that the ketone group is in the 1 position as shown (and not one of the two other theoretically possible monoketones having the keto group in the 2 or the 8 position) are furnished by the reaction with phosphorus pentachloride and its stability to heat. Proof that the product contains a "hexachlorocyclopentadiene rest" as part of its structure is shown by the fact that high temperature pyrolysis of the compound yields a substantial quantity of hexachlorocyclopentadiene.]

Reactions of the [decachlorotetrahydromethanoindeneone product] *$C_{10}Cl_{10}O$ ketonic compound* with the following: Phosphorus pentachloride, butyric anhydride, acetic anhydride; and the further reaction of the acetic anhydride reaction product with ethyl alcohol are described in the following examples.

EXAMPLE 5

Five parts of hydrated [decachlorotetrahydromethanoindeneone] *$C_{10}Cl_{10}O$ ketone* prepared as described in Example 4—purified by solution in methanol, precipitation with water and drying—were mixed with 21 parts of phosphorus pentachloride and the mixing heated for three hours at 125°–150° C. The resulting oily produce was cooled to 20° C. and drowned in water. The solid material which formed was filtered and water washed, then washed with methanol to dissolve any unreacted [decachlorotetrahydromethanoindeneone] *ketonic compound* which might be present, and dissolved in hot benzene. The benzene solution was mixed with a large excess of methanol to reprecipitate the product which was filtered, dried and then recrystallized from isopropanol. Two parts of *a* hexachlorocyclopentadiene dimer $C_{10}Cl_{12}$ were obtained. An infrared spectrogram prepared from this product was found to correspond to that shown as broken line A in Fig. 2 and to be identical with the dimer of hexachlorocyclopentadiene prepared by reacting hexachlorocyclopentadiene with aluminum chloride as described in JACS 71, page 954 (March 1949), in physical properties as well as its infrared spectrogram, shown as solid line B in Fig. 2.

EXAMPLE 6

One hundred parts of the [decachlorotetrahydromethanoindeneone] *$C_{10}Cl_{10}O$ ketone* hydrate prepared as described in Example 5 recrystallized from methyl alcohol and clarified with active carbon were mixed with 500 parts of freshly distilled acetic anhydride and the mixture was refluxed at 138° C. for six hours. The mixture was then vacuum distilled to remove the bulk of the unreacted acetic anhydride, leaving a slurry which was filtered. The crude solid amounted to 65 parts and melted at 138°–140° C. A portion of the crude solid was purified by recrystallizing from high boiling petroleum ether. The resulting purified acetate of [decachlorotetrahydromethanoindeneone] *$C_{10}Cl_{10}O$ ketone* had a melting point of 142° C. Upon heating with water or dilute acetic acid for two hours at 95° C., it was converted to a solid melting at 191°–194° C.

EXAMPLE 7

Fifty parts of [decachlorotetrahydromethanoindeneone] *$C_{10}Cl_{10}O$ ketone* hydrate prepared as described in Example 4 and purified by recrystallization were mixed with 312 parts of acetic anhydride and the mixture was refluxed for 3½ hours. After cooling the mixture to room temperature, it was drowned in several times its volume of cold water and allowed to stand overnight in contact with the water. A precipitate formed which was filtered and dried, producing a yield of 36 parts of crude product which melted from 238°–245° C. The crude product was recrystallized from isopropanol in the presence of decolorizing carbon, yielding a product melting at 231°–234°. A farther recrystallization from petroleum ether yielding a product melting at 251°–255° and apparently being a hydrated acetate of [decachlorotetrahydromethanoindeneone] *the $C_{10}Cl_{10}O$ ketone.* It had an infrared spectrogram corresponding to that shown as the broken line A in Fig. 3.

EXAMPLE 8

125 parts of [decachlorotetrahydromethanoindeneone] *$C_{10}Cl_{10}O$ ketone* hydrate purified by recrystallization from methanol, after clarification with decolorizing carbon were mixed with 700 parts of acetic anhydride and were heated at 135°–140° C. for 4½ hours. The reaction mixture was then cooled to room temperature, poured into 4000 parts of cold water and allowed to stand in contact with the water overnight to form and precipitate the hydrated acetate of [decachlorotetrahydromethanoindeneone] *the $C_{10}Cl_{10}O$ ketone.* The precipitate was then filtered, washed with water and dried, yielding 133 parts of crude acetate.

The crude acetate of [decachlorotetrahydromethanoindeneone] *the $C_{10}Cl_{10}O$ ketone* prepared above was mixed with 800 parts 95% ethyl alcohol and gently refluxed for 20 hours. During the initial portion of the heating, the mixture remained as a slurry. As reaction occurred, the solid alcoholate reaction product dissolved in the alcohol. After refluxing as described, the solution was evaporated to 350 ml., and cooled to 10° C. to bring about crystallization of the product. Upon filtration, 62 parts of product were obtained which upon purification by recrystallization from ethyl alcohol in the presence of decolorizing carbon yielded a purified dialcoholate of [decachlorotetrahydromethanoindeneone] *the $C_{10}Cl_{10}O$ ketone* melting at 123°–126° C. Its infrared spectrogram was made and is shown as the broken line A in Fig. 4.

The acetate of [decachlorotetrahydromethanoindeneone] *the $C_{10}Cl_{10}O$ ketone* was reacted with methyl alcohol in a manner similar to that described above yielding a methyl alcoholate reaction product melting at 146° C.; and with isopropanol yielding an isopropyl alcoholate melting at 127° C.

EXAMPLE 9

Thirty parts of purified [decachlorotetrahydromethanoindeneone] *$C_{10}Cl_{10}O$ ketone* hydrate were mixed with 90 parts of butyric anhydride. The mixture was refluxed for three hours at 190° C. The reaction mixture was then treated several times with boiling water, causing separation of an oily layer which finally solidified. 27 parts of solid product were obtained and were recrystallized from ethyl alcohol in the presence of decolorizing carbon yielding a monobutyrate of [decachlorotetrahydromethanoindeneone] *the $C_{10}Cl_{10}O$ ketone* melting at 173° C.

While the above describes the preferred embodiments of our invention, it will be understood that departures may be made therefrom within the scope of the specification and claims.

We claim:

1. The method of making a [decachlorotetrahydro-4,7-methanoindeneone] *ketonic compound having the empirical formula $C_{10}Cl_{10}O$* which comprises mixing hexachlorocyclopentadiene and sulfur trioxide at temperatures between about 35° C. and about 70° C. to form a reaction product thereof, and hydrolyzing the reaction product.

2. The method of making a [decachlorotetrahydro-4,7-methanoindeneone] *ketonic compound having the empirical formula $C_{10}Cl_{10}O$* which comprises mixing hexachlorocyclopentadiene and sulfur trioxide at temperatures between about 35° C. and about 70° C. to form a reaction product thereof, and drowning the thus formed reaction product in at least about 5 volumes of an aqueous drowning medium per volume of reaction mixture to hydrolyze the reaction product to the hydrate of the [decachlorotetrahydro-4,7-methanoindeneone] *ketonic $C_{10}Cl_{10}O$ compound.*

3. In a process for preparing [decachlorotetrahydro-4,7-methanoindeneone] *a ketonic compound having the empirical formula $C_{10}Cl_{10}O$,* the steps which comprise mixing hexachlorocyclopentadiene and sulfur trioxide at temperatures between about 35° C. and about 70° C. to form a reaction product thereof, drowning the thus formed reaction mixture in at least about 5 volumes of water, containing a small quantity of dissolved alkaline material, per volume of reaction mixture to hydrolyze the reaction product to the hydrate of the [decachlorotetrahydro-4,7-methanoindeneone] *ketonic $C_{10}Cl_{10}O$ compound.*

4. In a process for preparing [decachlorotetrahydro-4,7-methanoindeneone] *a ketonic compound having the*

empirical formula $C_{10}Cl_{10}O$, the steps which comprise mixing hexachlorocyclopentadiene and liquid sulfur trioxide at temperatures between about 35° C. and about 70° C. to form a reaction product thereof, drowning the thus formed reaction mixture in at least about 5 volumes of water containing a small quantity of dissolved caustic alkali per volume of reaction mixture to hydrolyze the reaction product to the hydrate of the [decachlorotetrahydro-4,7-methanoindeneone] *ketonic $C_{10}Cl_{10}O$ compound.*

5. In a process for preparing [decachlorotetrahydro-4,7-methanoindeneone] *a ketonic compound having the empirical formula $C_{10}Cl_{10}O$,* the steps which comprise mixing hexachlorocyclopentadiene and liquid sulfur trioxide at temperatures between about 35° C. and about 70° C. to form a reaction product thereof, drowning the thus formed reaction mixture in at least about 5 volumes of water containing a small quantity of dissolved caustic alkali per volume of reaction mixture to hydrolyze the reaction product to the hydrate of the [decachlorotetrahydro-4,7-methanoindeneone] *ketonic $C_{10}Cl_{10}O$ compound,* digesting the charge at a temperature between about 90° C. and about 95° C. for at least about 30 minutes, neutralizing the mixture at said temperature, thereafter digesting the charge at said temperature for at least about an additional 30 minutes, cooling the mixture to precipitate the hydrated product in crystalline form, and recovering the crystalline product.

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